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In Memory of Joan M. Daisey

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Foreword

This document compiles papers produced by staff and collaborators of the Indoor Environment Department at Lawrence Berkeley National Laboratory for presentation at the Indoor Air 2002 Conference, to be held June 30 – July 5, 2002 in Monterey, California. The Indoor Air Conference, held every three years, is the largest international conference on indoor air quality and was last held in the United States during 1981.

The compilation is dedicated to Dr. Joan M. Daisey who served as a Vice President of the organizing committee for the Indoor Air 2002 Conference until her death in February 2000. Joan was also the Department Head for the Indoor Environment Department. Please see the Dedication on the following page.

Acknowledgment

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DEDICATION TO JOAN M. DAISEY

This compilation of papers for the Indoor Air 2002 Conference, authored by staff at Lawrence Berkeley National Laboratory and by their collaborators, has been assembled to honor our beloved former colleague --Joan M. Daisey. Joan was a Vice President for the Indoor Air 2002 Conference until her death on February 29, 2000.

After receiving a Ph.D. in physical chemistry from Seton Hall in 1970, Joan Daisey's career started with teaching of undergraduate chemistry at Mount Saint Mary's College in New York state. She soon moved to a research position at NYU where, among other efforts, she studied the composition of aerosols in urban air and advanced gas chromatographic methods for the analysis of polycyclic aromatic hydrocarbons (PAH) in airborne particulate matter. Joan published more than 40 papers on the organic composition of atmospheric aerosols, including early work on their mutagenicity. She was Principal Investigator for the Airborne Toxic Elements and Organic Substances Study, an important study of non-criteria air pollutants which led to several significant papers on chemical composition and mutagenesis published in a book co-edited with Paul Lioy.

In 1986, Joan came to Berkeley to lead the Chemistry Group of Lawrence Berkeley National Laboratory's Indoor Environment Department (IED). Much of her research focused on organic chemicals in indoor air and on the characterization of environmental tobacco smoke. She had a remarkable ability to contribute importantly to a broad range of indoor and outdoor environmental research. In 1989, she was appointed as the Department Head for the IED. Under her leadership, the IED added a new research group on exposure and risk analyses and expanded an existing research effort, yielding a new group focusing on indoor air and pollutant transport. During this period, Joan also supported the expansion of an outdoor air quality research program at the Laboratory. At a time when indoor and outdoor air quality research efforts were often disconnected, Joan conceptualized these issues as a continuum and realized the value of coordinated programs of research.

The very high scientific standards of Joan's research were widely recognized. She was also clearly an innovator – always interested in the scientific frontiers in her field. As examples, she contributed substantially to early research on the transport of organic compounds via soil gas into buildings, chemical reactions among indoor pollutants, and the health effects of pollutant mixtures. Considering Joan's high scientific stature, open mind, and strong leadership and interpersonal skills, it was not surprising that she was sought as a leader for a range of scientific and professional committees. Foremost among these efforts was her service on the Science Advisory Board (SAB) of the U.S. Environmental Protection Agency. First appointed to the SAB in 1986, she was appointed co-chair of the Integrated Human Exposure Committee in 1993 and then chair a year later. Joan served as chair of the SAB's executive committee beginning in 1997 until her death. During her tenure, the SAB was very active—directly through the Executive Committee and via the various standing committees—resulting in 101 full reports, consultations, commentaries, and letter reports sent to the Agency with Joan's signature.

As colleagues, we recall Joan's friendship, mentoring, high energy level, and wit as much as her outstanding scientific accomplishments. The well being of colleagues was a priority for Joan. She enjoyed mentoring young scientists and she was able to motivate her young colleagues to go back and do a better and better job, without making them feel criticized. With her positive outlook and abundant energy, she was able to lift the spirits of those around her. Many of her colleagues became lifelong friends. Even in private farewells during the last days of her life, Joan was motivating, encouraging, and especially loving to her friends. Joan was a true inspiration to us all.

April 30, 2002
William Fisk, Richard Sextro, Nancy Brown, Al Hodgson, Hal Levin

I VENTILATION AND IAQ STUDIES

ENERGY AND INDOOR ENVIRONMENTAL QUALITY IN RELOCATABLE CLASSROOMS

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ABSTRACT
Relocatable classrooms (RCs) are commonly utilized by school districts with changing demographics and enrollment sizes. Four energy-efficient RCs were designed and constructed for this study to demonstrate technologies that simultaneously attempt to improve energy efficiency and indoor environmental quality (IEQ). Two were installed at each of two school districts, and energy use and IEQ parameters were monitored during occupancy. Two (one per school) were finished with materials selected for reduced emissions of toxic and odorous volatile organic compounds (VOCs). Each RC had two HVAC systems, alternated weekly, consisting of a standard heat-pump system and an indirect-direct evaporative cooling (IDEC) system with gas-fired hydronic heating. The hypothesized advantages of the IDEC include continuous outside air ventilation at ≥7.5 L s^{-1} per person, ~70% less cooling energy and efficient particle filtration. Measurements include: carbon dioxide, particles, VOCs, temperature, humidity, thermal comfort, noise, meteorology, and energy use. Preliminary IEQ monitoring results are reported.

INDEX TERMS
energy, indoor pollutants, ventilation, thermal comfort, school

INTRODUCTION
Energy efficiency in buildings is a key design consideration regulated by governmental agencies. Buildings also can be designed to explicitly improve indoor environmental quality (IEQ). Designs that achieve good IEQ can be expected to have beneficial effects with respect to occupant health, work performance and absenteeism (Fisk, 2000). This study is being conducted with the goal of quantifying and demonstrating technologies that simultaneously attempt to improve energy efficiency and IEQ using new relocatable (modular or portable) classrooms as the exemplary buildings. Relocatable classrooms (RCs) are particularly well suited for this demonstration because they are self-contained structures with dedicated HVAC systems and well-defined occupancies. In addition, a large number of these units are being constructed in California (CA). Here, we present the design, status and preliminary results for a field study of four new high-performance (HP) RCs that have been constructed and located at two schools. This study is a collaboration among Lawrence Berkeley National Laboratory (LBNL), Davis Energy Group (DEG), a RC manufacturer, and two CA school districts.
The use of RCs in California schools has increased dramatically in recent years due to an increasing student population and to state and federal mandates for class size reduction. An estimated 75,000 RCs are currently in place in California schools, and the numbers are

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increasing at a rate of at least 10,000 per year (EdSource, 2002, Waldman, 2001). The most common RC configuration is the two module 7.2-m x 12-m, 86-m^2 (960-ft^2) classroom, although a number of other styles, including a three module and two-story designs exist.

The HVAC system is a critical component of RC design from both energy and IEQ perspectives. Operating costs, electric demand, and other constraints influence HVAC design decisions such as equipment configuration, energy efficiency, and fuel source. The system also must be capable of providing adequate outdoor air ventilation since natural ventilation is often unfeasible and may be inadequate. The American Society of Heating, Refrigeration, and Air-Conditioning Engineers (ASHRAE) Standard 62-1999 (ASHRAE, 1999), as well as the State of CA Building Standards and Occupational Safety and Health Codes (CCR, 1995; CCR, Title 8) specify a minimum ventilation rate of 7.5 L s^{-1} per person in nonresidential buildings. Ventilation delivered at this rate will typically maintain indoor occupant-generated carbon dioxide (CO_2) at less than 1000 parts-per-million (ppm).

Standard CA RCs typically provide a ducted supply-air all-electric 10 SEER (Seasonal Energy Efficiency Ratio) compressor-based wall-mounted HVAC system with a direct return through the wall. These units are equipped with a movable damper that can be set to provide up to 25% of the delivered supply from outside air, providing ventilation sufficient to supply 7.5 L s^{-1} per person to about 30 occupants. In practice, the amount of outside air delivered to a classroom can be less due to fan on/off cycling or improper setup of the system. Additional HVAC characteristics affecting IEQ are mechanical noise and the efficacy of supply air filtration. Loud compressor and fan cycling noise is reported as disruptive to learning and results in reduced ventilation system usage. Air filtration, often minimal (10-20% ASHRAE dust spot efficiency) in many RC HVAC systems, is necessary to ensure low particulate matter (PM) concentrations indoors, especially in regions with elevated outdoor levels.

Considerable concern also has been generated regarding indoor levels of toxic and odorous volatile organic compounds (VOCs), including formaldehyde in classrooms, with special scrutiny placed on RCs. Careful selection of construction materials in combination with adequate ventilation can reduce baseline VOC concentrations and resultant occupant exposures to these chemicals.

METHODS

The field study phases include: school district and RC manufacturer recruitment; RC design specification and construction; RC installation at schools and instrumentation; field measurement and data collection during cooling and heating seasons; and data analysis. A concurrent energy simulation effort will enable extrapolation of collected energy data to predict energy savings by climatic zone and to conduct cost benefit analyses.

School districts and a RC manufacturer were simultaneously recruited for participation in the study. In order to test the RC designs in diverse climates, we wanted the schools to be located in two distinct regions: the CA Central Valley (extreme climate) and the San Francisco Bay Area (moderate climate). Several independent manufacturers in the CA modular classroom industry were approached. American Modular Systems (AMS, Manteca, CA) was willing to participate and had pending orders for numerous RCs from school districts in both of the desired regions. Through AMS, we secured agreements for placement of two HP RCs each at an elementary school in Modesto (MCS, Central Valley) and Cupertino (CUSD, SF Bay Area).

The HP RC design used in this study utilizes available energy efficient construction materials and methods including additional wall, floor, and ceiling insulation; ceiling vapor barrier, "Cool Roof" reflective roof coating, low-emissivity window glazing, and efficient (T8) fluorescent lighting (DEG, 2000). Each of the four study RCs are equipped with two HVAC systems: a standard 10 SEER heat-pump system, and an energy-efficient indirect/direct evaporative cooler (IDEC). The IDEC supplies continuous ventilation at ≥7.5 L s^{-1} per person even when heating or cooling is not required. Additionally, compared to the standard heat-pump system, it consumes about 70% less cooling energy, and as it has no compressor and a very quiet fan, the noise output from the system is much lower. Incorporated into the IDEC is an 85% efficient (AFUE) gas-fired hydronic space heating system and an inlet filter system with 65% ASHRAE dust spot efficiency (Apte et al., 2001). Both the IDEC and the heat pump system controls, as currently designed, require that the system be turned on in order to provide the required ventilation. In the case of the heat pump system, this action is tied to the temperature set point, such that outside air is only supplied when heat or cooling is needed.

In order to study VOC source reduction potential, one RC at each school received alternative low-VOC emitting wall panels, carpet, and ceiling panels (Hodgson et al., 2001 and 2002). Otherwise, all four RCs were constructed identically.

Table 1. IEQ and energy monitoring instrumentation in study relocatable classrooms.

Parameter	*Method*[1]	*Mfr. / Model*	*Location*[2]
Continuous:			
Carbon Dioxide	NDIR	Fuji / ZFP-9	I, O
Particle size, count	Laser counter	Met One / 237B	I,O
Relative Humidity	Capacitance	General Eastern/MRH	I, O,HPD, ID, TC, C
Temperature	Thermistor	YSI/44018 and 44303	I, O,HPD, ID, TC, C,
Air Velocity	Thermo-anemometer	Sensor Elect. & Meas. Equip. / HT412-2	TC
Sound Level	dB, A-wtd., Leq	Extech/407736.	C
Door open	Door sensor	LBNL	Door
Window position	LDP	JK Controls/LDP-500	
Wind speed, dir.	Anemometer	Davis/Industrial #7914	O
Electricity	Current transducer	CCS/Wattnode	HVAC, Lights, Total
Natural Gas	Gas meter	Equimeter/pulse output	IDEC Heating
Time-averaged:			
VOC	Multisorb GC/MS	LBNL[3]	I, O
Formaldehyde, acetaldehyde	DNPH HPLC UV detector	LBNL[3]/Waters	I, O
Thermal Comfort	ASHRAE 55-1992	LBNL[4]	***I (0.1m, 0.5m, 1.1m)***

[1]NDIR=non-dispersive infrared; multisorb=multisorbent tubes GCMS=gas chromatography/mass spectrometry; HPLC=high performance liquid chromatography, LDP = linear displacement potentiometer; A-wtd=A-weighted, Leq = equivalent noise level
[2]I=Indoors, O=Outdoors, HPD=Heat pump system diffuser, ID=IDEC Diffuser, TC=thermal comfort cart, C=Indoors@ 2.5m, center of RC, m=meters above floor
[3]See Hodgson *et al.*, 2001 [4]See ASHRAE, 1992

Two HP RCs were sited side-by-side at each of the schools prior to the fall 2001 semester. They were occupied and used by 3rd and 4th grade classes consisting of 20-25 students and one

teacher. During nine weeks of the 2001 summer/fall cooling season (August-October 2001) and currently during nine weeks of the heating season (January-March 2002) the two RCs at each school were simultaneously operated with either the standard heat pump or the IDEC unit, switching weekly. Each RC is instrumented to measure a range of IEQ and energy parameters (Table 1). Indoor and outdoor CO_2 concentrations are measured continuously. The particle counters measure particle number concentrations in six size ranges from 0.3 to 10 micrometers. Real-time data are stored as 6-minute averages to a central data acquisition system (CDAQS) operated continuously. During the study period, all four RCs are visited once a week (typically Tuesday or Thursday). Data stored on the CDAQS is retrieved. Integrated, 8- hour indoor and outdoor VOC and aldehyde samples are collected and later analyzed. A thermal comfort cart, designed and constructed at LBNL based upon ASHRAE 55-1992 thermal comfort standard (ASHRAE, 1992), is operated in three locations representative of occupant placement in each classroom during break periods. Observational information on HVAC usage, teacher and student clothing levels, and an inventory of cleaning and classroom supplies is collected. At the end of the school day, the system operation is switched from the standard HVAC to the IDEC, or visa-vers

RESULTS AND DISCUSSION

As of this writing, data analysis is in progress and only preliminary results from the IEQ monitoring are available. Thus, the results are observational rather than quantitative. The RCs were sited and commissioned at the beginning of the 2001 school year. The participating teachers and school custodians received training on the operation of the two HVAC systems and were briefed on field visit procedures and the schedule of weekly system switching. They were instructed to turn the HVAC system control on at the beginning of each school day. In order not to bias the teachers' behavior, we avoided discussing IEQ issues with the teachers and simply described the project as a study to test a new energy-efficient, quiet HVAC system.

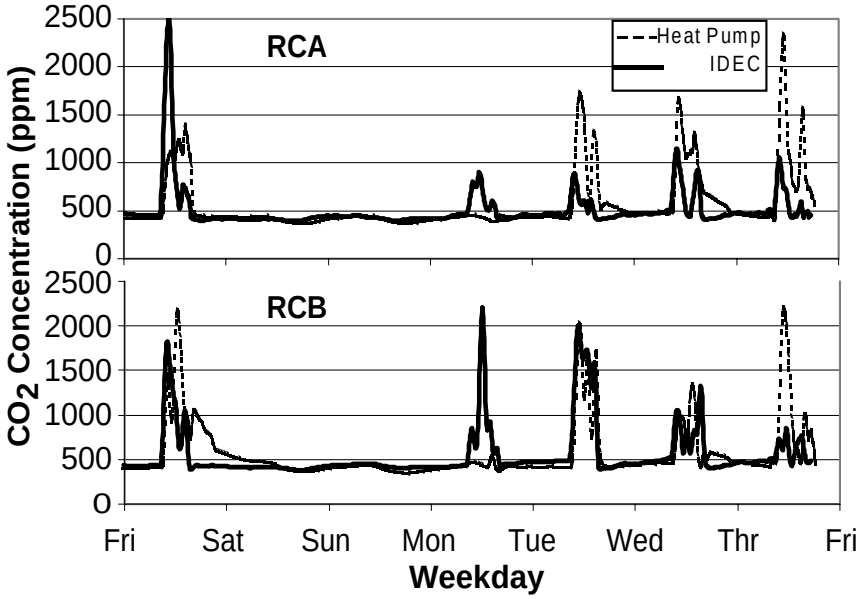

Figure 1. Indoor carbon dioxide concentrations in Cupertino study classrooms (RCA and RCB) during two consecutive weeklong cooling periods. The IDEC was operated during one week and the heat pump system was operated during the other.

Figure 1 displays weeklong indoor CO_2 concentrations in the two CUSD RCs for consecutive standard heat pump and IDEC weeks during the cooling season. No occupancy or HVAC

operation occurred during Saturdays or Sundays. The CO_2 data support a pattern verified by the energy and HVAC control data. The teachers' operation of the HVAC systems was not solely based upon cooling demand, and they did not always turn the IDEC on in the morning as instructed. During periods when the teachers did not provide ventilation, the CO_2 concentrations in the classrooms were elevated well above 1000 ppm, with peaks reaching about 2000 ppm, irrespective of the HVAC system. Note that during Monday of the IDEC week there was a field trip and both RCs were vacant. On Thursday of the IDEC week, our field staff was onsite and reminded the teachers to operate their systems the entire day. Indoor CO_2 concentrations on that day were well below 1000 ppm. Likewise, the CO_2 levels in RCA (but not RCB) reflect operation of the IDEC as instructed on Wednesday.

Table 2. Average (standard deviation) daytime indoor/outdoor particle-count concentration ratios in Cupertino study classrooms RCA and RCB during consecutive heat pump and IDEC weeks.

	Fri	Sat	Sun	Mon	Tues	Wed	Thr
	RCA						
IDEC	0.9 (0.2)	0.6 (0.0)	0.5 (0.0)	0.5 (0.1)	0.6 (0.1)	0.5 (0.0)	0.5 (0.1)
Heat Pump	1.0 (0.2)	0.7 (0.1)	0.6 (0.1)	0.7 (0.1)	0.7 (0.1)	0.6 (0.1)	0.6 (0.1)
	RCB						
IDEC	0.6 (0.1)	0.5 (0.1)	0.2 (0.1)	0.3 (0.1)	0.5 (0.1)	0.3 (0.0)	0.3 (0.0)
Heat Pump	0.8 (0.3)	0.4 (0.0)	0.3 (0.0)	0.4 (0.1)	0.5 (0.1)	0.6 (0.1)	0.4 (0.1)

Average (±standard deviation) daytime indoor particle number concentrations in the same classrooms during the same weeks were 64 ±55 x 10^6 and 47 ±45 x 10^6 particles m^{-3} for RCA and RCB, respectively. The simultaneous outdoor particle concentrations were 98 ±87 x 10^6 particles m^{-3}. The large standard deviations of particle concentrations reflect the variability of daytime particle concentrations. Table 2 presents daytime (8:00-15:00) indoor to outdoor (I/O) cumulative particle count ratios (sum of all size bins). The I/O ratio was highest on both Fridays. This is consistent with the elevated CO_2 levels (Figure 1) and is most likely due to the combination of low ventilation and re-suspension of particles by the occupants. During Monday through Thursday on both IDEC and standard heat pump weeks, the I/O ratios were well below unity, but consistently lower during IDEC operation. On Thursday during proper IDEC operation, the ratio was about 0.5 in RCA and 0.3 in RCB. Overall it appears that particle infiltration from outdoors or indoor particle re-suspension was less in RCB.

Preliminary findings from the VOC measurements made in the RCs when they were operating with the IDEC system are presented elsewhere (Hodgson et al., 2002).

As discussed above, the teachers' patterns for operation of their HVAC systems directly influenced classroom IEQ parameters during the school day. As noted, both systems, as currently designed, must be turned on in order to provide the required ventilation. The heat pump thermostat does provide a "Fan Only" setting, but the teachers may prefer not to use it due to fan noise. The IDEC control requirement is only that it be on during occupancy since its fan provides continuous 100% outside air when it is turned on. In some cases the decision to not turn the systems on is based upon a desire to save energy. For example, to save energy one teacher in the study regularly opened the RC windows during the morning instead of running the HVAC. The resulting indoor CO_2 levels often rose above 1000 ppm indicating that windows alone may not have provided adequate ventilation for the 21 occupants.

Data analyses will include multivariate regressions measuring operational parameters against the IEQ metrics, in order to identify the effectiveness of the IDEC system. Thermal comfort data will be analyzed to identify the extent to which the IDEC system is able to provide a thermally acceptable environment. Energy analyses will provide a basis for estimating the cost effectiveness of this HP RC package including the IDEC system as an alternative to the standard RC designs currently in use.

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VENTILATION EFFICIENCIES OF A DESK-EDGE-MOUNTED TASK VENTILATION SYSTEM

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ABSTRACT
In chamber experiments, we investigated the effectiveness of a task ventilation system with an air supply nozzle located underneath the front edge of a desk and directing air toward a heated mannequin seated at the desk. The task ventilation system provided outside air, while another ventilation system provided additional space cooling but no outside air. Test variables included the vertical angle of air supply (-15° to 45° from horizontal), and the supply flow rate of (3.5 to 6.5 L s^{-1}). Using the tracer gas step-up and step-down procedures, the measured air change effectiveness (i.e., exhaust air age divided by age of air at the mannequin's face) ranged from 1.4 to 2.7, which is higher than typically reported for commercially available task ventilation or displacement ventilation systems.

INDEX TERMS
Ventilation rates and strategies, Offices, Improved IAQ practices and technologies, Perceived air quality

INTRODUCTION
Numerous studies have found that increased outside air ventilation rates in buildings are associated with reduced sick-building syndrome (SBS) health symptoms and with improvements in perceived air quality (Seppanen et al., 1999). Laboratory studies suggest that worker performance may improve with higher ventilation rates (Wargocki et al., 2000).

In general, increased ventilation rates will increase building energy use; therefore, technologies or practices that bring about the benefits of increased ventilation rates without energy penalties are highly desirable. One general approach for obtaining the benefits of increased ventilation rates without actually increasing the quantity of outside air supply or the associated energy use is to supply outside air in a manner that preferentially ventilates the breathing zone.

To quantify the benefits of such air supply methods, we use the air change effectiveness (ACE). The ACE is a measurable parameter used to compare the effective ventilation rate at the breathing zone to the effective ventilation rate that would occur throughout the building with thoroughly mixed indoor air at the same rate of outside air supply. ASHRAE (1997), defines the ACE based on ages of air as follows

$$ACE = \frac{\tau_n}{\tau_{avg}} \quad (1)$$

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where τ_n is the nominal ventilation time constant and τ_{avg} is the average age of air at the breathing zone. The average age of air at a location is the average time elapsed since air at that location entered the building. τ_n equals the average age of air exiting the building which is identical to the indoor air volume divided by the rate of outside air supply. The practical interpretation is that ACE equals the effective ventilation rate at the location where people breathe divided by the ventilation rate that would occur throughout the ventilated space with perfect mixing. Consequently, we seek ventilation technologies with ACEs that are as high as possible.

In the U.S., traditional ventilation systems supply a mixture of outside and recirculated air in high velocity jets so that the indoor air in rooms is often well mixed and the ACE is approximately unity (Fisk et al. 1992, 1997; Olesen and Seelen 1992; Persily 1986; Persily and Dols 1989). Task-ambient conditioning (TAC) systems are a ventilation technology with the potential for improved ACE. TAC systems may supply air from the floor, desk, or partitions and enable occupants to adjust the supply flow rate, direction, or temperature so that thermal conditions can be tailored to meet the individual's requirements. Most TAC systems supply a mixture of outside and recirculated air in relatively high velocity jets. The mixing caused by the air recirculation and the jet-induced air motions prevent ACE values from substantially exceeding unity. However, in experiments using these systems to supply only outside air, ACE values were sometimes significantly above unity, but generally still below 1.3 (Faulkner et al. 1993, 1993b, 1999; Fisk et al. 1991)

In prior experiments (Faulkner et al. 1999), one TAC system designed to supply only outside air had relatively high values of ACE. This system supplied air horizontally toward the seated worker's torso from the underside of the desk top, with nozzles located approximately 70 cm from the occupant, and resulted in ACE values up to 1.4. The system could also supply air vertically toward the face from the front edge of the desk, leading to ACE values up to 1.9, but these very high ACE values only occurred with the mouth and nose located precisely within the supply jet.

Our current research seeks to identify air supply methods that provides consistently high values of ACE, for example by moving the outside air supply closer to the occupant and supplying air in low velocity jets. It is imperative that these air supply methods do not degrade thermal comfort, cause unacceptable drafts, or irritation at the face and eyes due to high air velocities. A similar research effort is underway at the Danish Technical University, with some promising results (Cermak and Majer 2000).

METHODS

The experimental system, illustrated in Figure 1, has a heated thermal mannequin seated at a desk, with a personal air supply of 100% outside air located beneath the keyboard. The desk and mannequin are located within a nearly airtight experimental room with a volume of 27 m^3, located within a thermally conditioned, nearly constant temperature, laboratory.

For reliable evaluations of the Task Ventilation system, the experimental room must have realistic sources of indoor air motion and mixing. In this facility, air motion is driven by the heated mannequin, the heat release and fan of a personal computer, heat release from a computer monitor and overhead light, and by a recirculating air stream. The air supply jet of the Task Ventilation system provides additional indoor air movement. The mannequin is

wrapped in electrically resistant media located beneath clothing, and the voltage supplied to the media was adjusted to produce a total rate of sensible heat generation of 75 W, typical of the sensible energy release of an adult office worker. The energy consumed by the personal computer, monitor, and overhead light are 50 W, 62 W, and 29 W, respectively. The recirculating air stream (100% recirculated air) has an air flow rate of 33 L s^{-1} equivalent to 4.4 indoor air volumes per hour, and represents the airflow that would typically be used to heat or cool rooms. Air exited the experimental room from the recirculated airstream. The exhaust flow rate was adjusted manually, using a damper, to keep the room pressurized by ~ 1.5 Pa.

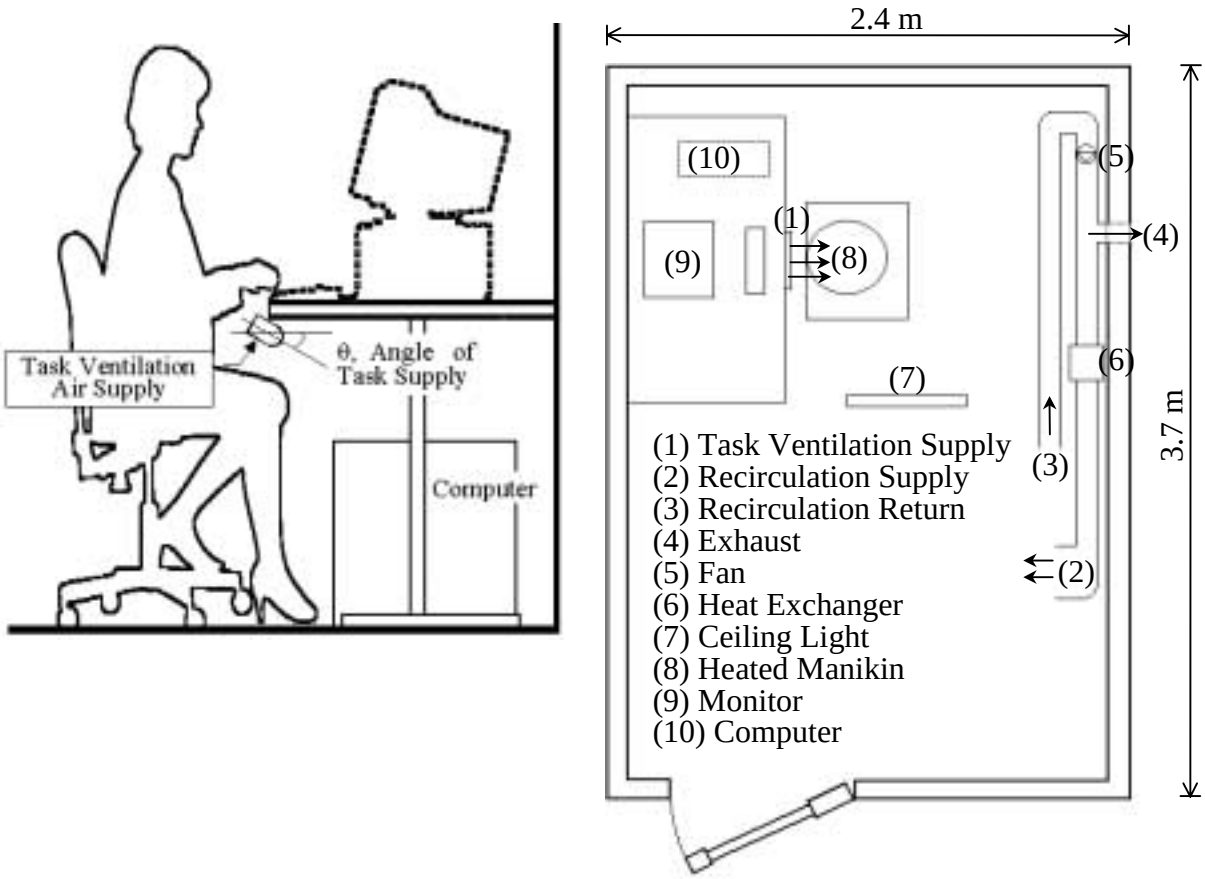

Figure 1. Side view of mannequin at desk in chamber and plan view of chamber with equipment identified.

The air supply nozzle of the Task Ventilation system was located beneath the front edge of the desk, about 10 cm from the mannequin. It was constructed from a 3.8 cm diameter PVC pipe with a slot, 3.8 cm high and 30.5 cm long. The slot was filled with a 3.2 mm per side hexagonal flow straightener. The angle of the nozzle was adjustable so that air could be directed from -15° (15° downward from horizontal) to +45° (see Figure 1).

Our prior experiments (Faulkner et al. 1999) and those of Cermak and Majer (2000), plus airflow visualization studies, indicated that outside air supplied at a low velocity near the waist, will be entrained in the thermal plume flowing upward around the mannequin and thus,

be transported to the region of the mouth and nose. The exit area of the nozzle is 116 cm^2, resulting in a nominal air supply velocity of 0.3 m s^{-1} with a supply flow rate of 3.5 L s^{-1}. With such a low supply velocity, the air supply jet, when at room temperature, can just be sensed if one places their hand between the nozzle and the mannequin. Velocities measured about 8 cm in front of the upper chest were typically less than 0.1 m s^{-1}. The turbulence of the supply jet is reduced using a honeycomb flow straightener, because the theoretical work of Cermak and Majer (2000) suggests improved task ventilation system performance with reduced turbulence. A small heat exchanger located in the supply pipe for the Task Ventilation enabled the supply temperature to be reduced to as low as 17 ^oC, about 5-6 ^oC less than the ambient room temperature.

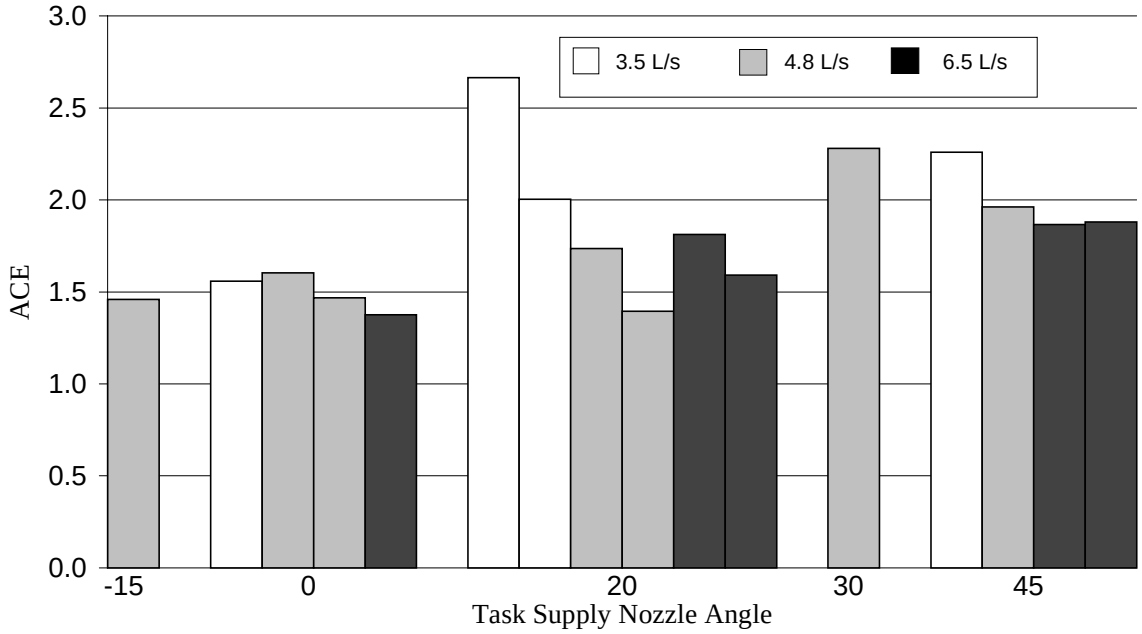

Figure 2. ACE values at outside air flow rates of 3.5, 4.8 and 6.5 L s^{-1} and task supply nozzle angles from -15° to 45°.

The ACE was measured using well-established tracer gas stepup or decay procedures (ASHRAE 1997). During tracer stepups, sulfur hexafluoride (SF_6) tracer gas was injected in the Task Supply pipe at a location upstream of the supply fan to increase mixing of the tracer within the airstream. The injection rate was maintained at a constant rate with a mass flow controller. Tracer gas injection continued for 3.5 to 9.5 hours, dependent upon the outside air flow rate. At the end of the stepups, SF_6 concentration in the room's exhaust airstream was steady within the precision of our measurement system. For tracer gas decays, the tracer gas injection at the end of a stepup was stopped, allowing indoor SF_6 concentrations to decay.

A quadrapole mass spectrometer with a multi-port valve was used to measure the tracer gas concentrations at 14 locations, every 2 minutes, around the mannequin and in the chamber. In this paper we concentrate on the measured concentrations at the mannequin's nose and in the

chamber exhaust stream. The mass spectrometer was calibrated before each experiment with 20 calibration gases, and has a very linear calibration curve.

RESULTS & DISCUSSION

A sample of results is provided in Figure 2. Measured values of ACE range from 1.4 to 2.7. With a positive air supply angle, the ACE was typically 1.7 or higher. These values of ACE are higher than typically measured in our prior studies of commercially available task ventilation systems and are also higher than typically reported for displacement ventilation systems. In a set of four repeat tests, the ACEs differed, on average, by 0.2. These results suggest that improved air supply technologies for task ventilation have a significant potential to improve IAQ at the breathing zone without increasing energy use.

Figure 2 illustrates a couple relationships that warrant comments. First, at a given flow rate, as the nozzle angle increases, the ACE increases also. As the nozzle angle increases so that it points more toward the mannequin's face, it provides upward momentum to carry the fresh outside air toward the breathing zone. There most likely is an optimum angle, possibly 30° to 45° for the nozzle angle to deliver the maximum ACE. This optimum angle may be dependent upon the flow rate.

The second observation is that ACE values tend to be greater at lower flow rates. This is important from an energy standpoint, as energy increases as outside air flow rates increases. Also, lower flow rates will less likely cause a problem with drafts. Again, there must be an optimum flow rate, since allowing the flow rate to go to zero will not increase the ACE indefinitely.

Additional research is underway to further optimize task ventilation air supply technologies and to assess the effects on comfort. We also plan to measure the ACE values experienced by people, as opposed to a thermal mannequin. We expect some reduction in ACE due to the movements of real people.

CONCLUSIONS

We have measured ACE values, 1.4 to 2.7, from a task ventilation system that consistently shows better ventilation effectiveness than our previous studies of task ventilation. These high ACE values have been achieved at relatively low flow rates and thus should provide energy savings as compared to conventional overhead ventilation systems. Based on limited temperature and air velocity measurements, we believe that thermal comfort has not been compromised to achieve high ACE values.

ACKNOWLEDGEMENTS

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A PRIORITY AGENDA FOR ENERGY-RELATED INDOOR ENVIRONMENTAL QUALITY RESEARCH

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ABSTRACT
A multidisciplinary team of IEQ and energy researchers is working together to define a program of priority energy-related IEQ research. This paper describes the methods employed, ten high priority broad research and development (R&D) goals, and 34 high priority R&D project areas linked to these goals.

INDEX TERMS
Energy, IEQ research agenda, priorities

INTRODUCTION
Indoor environmental quality (IEQ) and building energy use are both strongly influenced by a building's design, construction, operation, and maintenance, by the activities of occupants, and by outdoor environmental conditions. Consequently, energy-efficiency measures may degrade IEQ, improve IEQ, or be IEQ neutral. Similarly, IEQ improvement measures may increase or decrease energy consumption or be energy neutral.

Given the many interactions between building energy performance and IEQ, these two issues must be addressed and researched in a coordinated manner. The U.S. Department of Energy (DOE) recognized the linkage of IEQ and energy use in the late 1970s and has since supported a modest size but important program of energy-related IEQ research. With the growing evidence that large health and productivity gains could be attained from practical improvements in IEQ, a group of state energy organizations represented by the Association of State Energy Research and Technology Transfer Institutions (ASERTTI) has recently expressed their support for an expanded and coordinated program of energy-related IEQ

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research. As a first step, ASERTTI members and DOE are jointly supporting the development of an agenda for priority energy-related IEQ research. Among ASERTTI members, the California Energy Commission, with assistance from the California Institute for Energy Efficiency, has taken a lead role in supporting the development of this research agenda. This paper describes the draft priority R&D agenda.

METHODS

In conjunction with the sponsors, a scope for the R&D agenda was established. The agenda was to define the highest priority research needs pertaining to the interrelationships among IEQ conditions (excluding lighting and acoustics), health, occupant satisfaction, worker performance, building energy use, and the building systems and practices affecting energy use. The scope was to include residences, non-industrial commercial buildings (excluding health care buildings), and schools and to consider the design, construction, and operation phases of buildings. The agenda was to include a balanced portfolio of R&D products, ranging from new knowledge to applied products for implementation by industry.

A multidisciplinary "core team" of leading IEQ and energy professionals (see list of authors) is developing this agenda. Core team members were approved by a steering committee of the project's sponsors. An advisory team representing the buildings' industry and other stakeholders will be reviewing and commenting on the draft agenda presented in this paper.

To provide a framework for the identification of priority R&D needs, the following activities were undertaken:

1. Given the focus on *energy-related* IEQ research, the processes that link IEQ with energy use, and the IEQ conditions closely linked to energy use, were tabulated.
2. The core team reviewed several existing documents that propose IEQ research agendas (DOE, 2000; EPA, 2000; Fisk, 2000; Mendell et al., 2002; Wyon, 2000)
3. To enable the core team to identify R&D needs that are not being adequately addressed in existing programs, the existing IEQ research activities of U.S. federal agencies, ASHRAE, Air Conditioning and Refrigeration Technology Institute, and the California Air Resources Board were investigated and summarized.
4. The following criteria were identified for qualitative judging of the priority of proposed R&D activities: potential health benefits; potential comfort or odor benefits; potential productivity benefits; potential energy benefits or strength of the linkage to energy; need for expanded *public* support of R&D; potential for R&D success; expected overall benefit to R&D cost ratio; likelihood that R&D results will be used; extent to which project advances science or technology.

With this groundwork completed, the core team members developed a set of ten priority R&D goals. These broad goals reflect a consensus among the core team members and convey what the core team wants to have accomplished via this expanded program of research. For each broad goal, the core team then identified 7 to 10 priority research project areas. Via a process of discussion and voting, the core team selected the top 34 and, among them, the top 22 priority R&D project areas. Text is currently being developed to describe and justify the top 34 priorities.

Table 1. Priority R&D goals and project areas related to advancing IEQ knowledge

Goa1 1. Identify IEQ problem areas and opportunities to improve IEQ by filling critical gaps in information about IEQ and about IEQ-influencing characteristics within existing buildings.

Project Areas:

- Characterize ventilation rates and IEQ conditions in energy efficient and conventional new housing. **Top22**
- Characterize ventilation and air-flow performance in existing buildings as a function of region, building type, HVAC system type. **Top22**
- Characterize IEQ in small (< 2500 m^2) commercial buildings. **Top 22**

Goal 2. Advance knowledge of how to improve IEQ, health, comfort and productivity by improving our understanding of the relationship of these outcomes with building ventilation, infiltration, and uncontrolled airflows.

Project Areas:

- Compare health outcomes among students in schools with very high and very low ventilation rates. Top22
- Investigate the influence of ventilation (natural and mechanical) on concentrations of contaminants from a diverse range of contaminant sources. Top22
- Relate ventilation rates in residences to health symptoms and to satisfaction with IAQ.
- Quantify the influence of outside air ventilation rates on prevalences of communicable respiratory diseases.

Goal 3. Advance knowledge of how to improve health, comfort and productivity by improving our understanding of the relationship of these outcomes with IEQ conditions and associated building characteristics (other than ventilation, infiltration, and uncontrolled airflows which are addressed in goal 2).

Project areas:

- Quantify the relationships between indoor chemical and biologic contaminants of greatest current concern and health effects on occupants. Top22
- Identify the health, energy, comfort, and productivity impacts of access to fixed and operable windows. Top22
- Characterize the effects of thermal comfort parameters on health and comfort in commercial and institutional buildings, including comparisons between air-conditioned and naturally ventilated buildings. Top22

Goal 4. Advance knowledge and models of how important IEQ conditions depend on characteristics and operation of buildings, providing as basis for improvements in building design, operation and maintenance and associated guidelines, standards, and codes.

Project areas:

- Characterize the impact of air flows, pressure differentials, ventilation rates, and HVAC system performance on contaminant transport and indoor humidity, and on related IEQ problems in small commercial buildings located in humid climates, and develop associated remediation measures. Top22
- Based on health and comfort effects, determine, as a function of climate, if the use of economizer cycles can enable a reduction of minimum ventilation rates.
- Quantify the IEQ and energy impacts of building/HVAC maintenance and space cleaning.

Table 2. **Priority R&D goals and project areas related to developing IEQ tools, technologies,**

Goal 5. To enable energy-efficient improvements in IEQ, characterize indoor pollutant sources and develop pollutant source reduction measures. **Project areas**: For various climates, develop best ventilation and insulation practices for crawl spaces and attics to minimize moisture problems and energy waste. Top22 Characterize the dependence of microbiologic growth on surfaces within buildings and HVAC systems upon the materials used, airflows, humidities, moisture contents, condensation, and soiling; and develop scientifically-documented strategies for material selection, design, and construction of building envelopes and HVAC systems that minimize risk of microbiologic contamination, while maintaining energy efficiency. Top22 Determine the pollutants, with their source strengths, emitted from common energy consuming office equipment such as copiers, printers, computers, etc.
Goal 6. Improve the IEQ performance of environmental control technologies and systems and develop new innovative technologies and systems for environmental control, including technologies and systems controlled by the building's occupants. **Project areas**: Develop new and innovative HVAC system designs that provide improved IEQ at minimum life cycle cost. Top22 Identify the components and features of current HVAC technologies posing risks, in actual use, of microbiologic contamination and dissemination. Top22 Develop and evaluate strategies for micro-zoning while providing individual control, with a focus on spatial and temporal variability in IEQ conditions, on occupant interactions with the building [including its systems and environment] and on the resulting effects on health, comfort, productivity, and building energy use.
Goal 7. To facilitate more efficient prevention or diagnosis of IEQ problems, develop practical measurement tools and diagnostic procedures for IEQ conditions of particular concern. **Project areas**: Develop inexpensive instruments for rapid and sensitive identification and measurement of indoor pollutants of concern. Top22 Develop diagnostic protocols and tools for failures of HVAC that deteriorate IEQ and energy efficiency. Top22

Table 3. Priority R&D goals and project areas related to facilitating the use of existing IEQ knowledge.

Goal 8. Develop IEQ performance metrics, guidelines, and standards that guide building design, construction, operation, and maintenance.

Project areas:

- Based on a cost/benefit analysis, determine the appropriate minimum ventilation rates in building codes. Top22
- Combine current scientific and practical knowledge on health effects of building design operations and maintenance, including ventilation, into guidelines for best building practices. Top22
- Use existing knowledge to develop recommended maximum pollutant concentrations for pollutants and pollutant mixtures of particular concern. Top 22
- Integrate materials, IAQ, IEQ knowledge and appropriate weighting factors into the LEEDS Rating System for Green Buildings. Top22

Goal 9. Develop and evaluate strategies and instruments to disseminate critical IEQ information, and decision-making tools that enable building professionals to improve IEQ in an energy efficient manner.

Project areas:

Identify key decision makers for IEQ and energy efficiency in buildings through the building life cycle, and develop effective information dissemination and decision-making tools for IEQ and energy efficiency specifically targeted to these individuals.

Create a consumer-report style document that ranks the performance IEQ related building products in use.

Develop a database of costs and economic benefits of energy efficient practices and technologies for improving IEQ.

For design professionals and code officials, develop and deliver education programs in HVAC and building science related to best practice for IEQ.

Goal 10. To stimulate improvements in IEQ, develop financial incentives and remove barriers.

Project areas:

Develop life cycle cost analysis methods that include human and energy effects of IEQ. **Top22**

Develop financial incentive tools to stimulate energy-efficient or energy-neutral improvements in IEQ in commercial and institutional buildings, such as: model lease language; marketing strategies a la the LEEDS rating system; model IEQ insurance policies; and model IEQ/energy efficiency disclosure requirements linked to sales. **Top22**

Document the full human and environmental cost of least-cost decision-making including health cost, energy consequences, productivity losses, and waste produced. **Top22**

Better quantify the productivity costs and investigation and remediation costs of SBS problems.

Develop financial incentives for the design and construction team that reward high IEQ and energy performance.

RESULTS

Tables 1-3 list the 10 priority goals organized into three goal categories: 1) Advancing IEQ knowledge as needed to better address IEQ in decisions about building design, operation, and maintenance and to enable improved guidelines, standards and codes; 2) Developing tools, technologies, and practices that will facilitate the diagnosis and energy-efficient prevention or remediation of IEQ problems, and 3) Facilitating the implementation of existing knowledge about how to maintain a high level of IEQ. The top 34 priority research project areas are listed under the most closely related goals, with the top 22 priorities labeled. Some priority goals and some priority research project areas do not fit unambiguously within a single category; thus, we have used our judgment to select the best category. In particular, some priority "*R&D*" areas call for facilitating the implementation of existing knowledge, when the existing knowledge about an issue is incomplete. Consequently, facilitating existing knowledge and development of related new knowledge should proceed in parallel.
and practices.

DISCUSSION

The identified R&D priorities reflect a strong need to better characterize IEQ conditions and to better understand how people are affected by IEQ conditions and by the related building characteristics or practices. Studying the effects of building characteristics and practices on IEQ conditions, which provides the basis for energy efficient and effective IEQ control measures, was also considered a priority. Advancements in IEQ-related tools, technologies, and practices, primarily those related to indoor pollutant sources and to HVAC, were identified as priorities. Despite the gaps in existing knowledge, the core team believes that existing knowledge about how to improve IEQ is underutilized; thus, many of the priority "research" project areas identified (Table 3) focus on facilitating the increased use of existing knowledge. Some readers may not characterize these activities as true scientific research, but the message from the core team is that these activities deserve a high priority.

Consistent with the focus on "energy-related" research priorities, building ventilation and HVAC are prominent in the agenda. The agenda also specifically calls for research on residences, *small* commercial buildings, and schools because these types of buildings have been underrepresented in prior research. Research related to moisture and microbiological problems, particularly within hot and humid climates, are also prominent within the agenda. Many of the priority needs identified apply for both existing buildings and new construction.

It is important to recognize that this agenda has been tailored to match the missions and R&D capabilities of the sponsors; hence, many important research needs may be omitted from the agenda, e.g., studies of the mechanisms underlying IEQ-related health effects. Additionally, very important areas of research currently being addressed by sponsors may not have ended up within the list of highest priorities, solely because our goal was to identify R&D needs that are not being adequately addressed in existing R&D programs.

How should sponsors and researchers use this agenda? We recommend that they use the final version of this agenda, as it will reflect the comments of an advisory team. This final agenda will be made available at the following address < http://eetd.lbl.gov/ied/ied.html> after

approximately July 15, 2002. We believe that many meritorious research efforts will be consistent with the priorities in this agenda. Specific research efforts may contribute to multiple goals and research project areas, or be more narrowly focused. Multidisciplinary research approaches are strongly encouraged. Finally, because it is likely that the core team has overlooked some critical research needs, we caution that sponsors should not exclude projects simply because they do not align with the priorities in this agenda.

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EFFECT OF OUTSIDE AIR VENTILATION RATE ON VOC CONCENTRATIONS AND EMISSIONS IN A CALL CENTER

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ABSTRACT
A study of the relationship between outside air ventilation rate and concentrations of VOCs generated indoors was conducted in a call center. Ventilation rates were manipulated in the building's four air handling units (AHUs). Concentrations of VOCs in the AHU returns were measured on 7 days during a 13-week period. Indoor minus outdoor concentrations and emission factors were calculated. The emission factor data was subjected to principal component analysis to identify groups of co-varying compounds based on source type. One vector represented emissions of solvents from cleaning products. Another vector identified occupant sources. Direct relationships between ventilation rate and concentrations were not observed for most of the abundant VOCs. This result emphasizes the importance of source control measures for limiting VOC concentrations in buildings.

INDEX TERMS
Ventilation, VOCs, Formaldehyde, Emission factors, Office buildings

INTRODUCTION
The importance of adequate ventilation in office buildings is supported by a number of studies that have investigated the association of ventilation rates with human responses as reviewed by Seppanen et al. (1999). Many of the included studies found that ventilation rates below 10 L s^{-1} per person were associated with one or more adverse health or perceived air quality outcomes. Ventilation rate increases above this rate were found in some studies to decrease symptom prevalence or to improve perceptions. These improvements were presumably due to reductions in concentrations of indoor contaminants, such as volatile organic compounds (VOCs). Mass balance considerations directly relate ventilation and concentrations of contaminants generated indoors. However, in practice the relationship is complicated by a number of potential factors affecting ventilation rates, air mixing, source strengths and removal mechanisms (ibid.). Ventilation additionally influences chemical reactions among indoor contaminants (Weschler and Shields, 2000). Thus, the effectiveness of ventilation for controlling indoor VOC concentrations may vary widely within and among buildings.

This study was conducted in support of an investigation of ventilation rate and worker productivity in a call center (Fisk et al., 2002). The primary objective was to determine how concentrations of VOCs generated indoors were affected by manipulated ventilation changes.

METHODS
The study was conducted in a health maintenance organization call center located in the San Francisco Bay Area. The building, constructed in 1998, had two floors, a total floor area of 4,600 m^2, and a worker density of ~6.3 persons per 100 m^2 (~290 persons). The building was ventilated and conditioned by four air handling units (AHUs 1-1, 2-1, 2-2 and 2-3) located on the rooftop. Each AHU served a defined interior zone (840-1,460 m^2), although there likely

was considerable air mixing among zones. AHU 2-2 served the entryway, lounge, and offices for management staff and meetings. Advice nurses and support staff occupied the other zones. Each AHU had an economizer control system. Equipment was added to enable manipulation and measurement of outside air ventilation rates as described elsewhere (ibid.). For the three AHUs serving advice nurses, three fixed damper positions were selected to provide periods of low, medium, and high ventilation rates. The damper positions for the low period were fixed to provide near the code-minimum outside air supply rate of 76 L s^{-1}/100 m^2. The damper positions for medium and high periods were selected to provide about 2- and 4-times the code minimum. The dampers in AHU 2-2 were fixed in one position. Using these methods, periods of ventilation were scheduled over 13 weeks (July 28 through October 24, 2000) in one of four modes: low, medium, high, and economizer mode (ibid.). CO_2 concentrations were measured on a 7-min cycle in all supply and return air systems and outside air.

Sampling for volatile organic compounds (VOCs) and aldehydes was conducted on weeks 2, 3, 6-8, 11 and 13. Air samples were collected from the return air ducts of the four AHUs and outside air on Tuesdays. Sampling began at 9:00 and ended at 15:00 to include the period of maximum occupancy. VOC samples were collected on sorbent tubes and analyzed for 49 compounds using thermal desorption GC/MS. Formaldehyde and acetaldehyde samples were collected on treated cartridges and analyzed using HPLC. The methods are described elsewhere (Hodgson et al., 2000). There were 6 field blanks for VOCs and aldehydes and 12 VOC sample pairs. Concentrations of individual VOCs and aldehydes (ppb; 25° C, 1 torr) and CO_2 (ppm) were determined for each zone and day of sampling. Emission factors (µg m^{-2} h^{-1}) were calculated by simple mass balance using the concentration data, the outside airflow rates in each AHU and the floor areas of the zones. Seven of the 28 samples were excluded from this analysis because sampling started shortly after an airflow rate was changed and the zone was not near steady-state conditions during a sufficiently long portion of the interval. The emission factors of the 10 predominant compounds plus CO_2 in the 21 samples collected at steady state were subjected to principal component (PC) analysis to identify groups of co-varying compounds based on common source types. The software was StatView for Windows (Ver. 5.0.1, SAS Institute Inc.). The varimax rotation method was employed.

RESULTS AND DISCUSSION

The normalized airflow rates (L s^{-1}/100 m^2) are presented in Figure 1. By design, AHU 2-2 flow rates remained relatively constant (<25% variation). There was an approximate 5-, 7- and 3-fold variation over the 7 days between the low and high rates in AHUs 1-1, 2-1 and 2-3, respectively. The building average flow rates ranged between 118 and 285 L s^{-1}/100 m^2 over the 7 days. These are equivalent to 1.5-3.8 times the code-minimum outside air supply rate.

Since the primary focus was on contamination sources associated with the building and occupants, indoor minus outdoor (In-Out) VOC and CO_2 concentrations were calculated. This eliminated outdoor motor vehicle contamination from consideration. Of the 51 analytes, which included abundant, toxic and odorous compounds, only 10 had median In-Out concentrations for the 28 samples in excess of 0.5 ppb (Table 1). With the exception of formaldehyde and acetaldehyde, the In-Out median values for these 10 exceeded their median outdoor concentrations by a factor of two or more. The concentrations of the predominant VOCs were generally consistent with central tendencies and maximum values reported in cross-sectional surveys of U.S. office buildings (Daisey et al., 1994; Girman et al., 1999).

There were at least 6-fold variations in individual VOC concentrations with space and time over the 7 days. Sampling in week 7 was preceded by four days at the medium ventilation rate in all zones. At this condition, the In-Out concentrations of CO_2 and all VOCs except acetaldehyde and d-limonene varied by less than a factor of three among all four zones.

Table 1. Concentrations and emission factors of most abundant VOCs and CO_2.

	Concentration (ppb)				Emission Factor[a] ($\mu g\ m^{-2}\ h^{-1}$)	
Compound	Outdoor Median	In-Out Median	In-Out Range	Precision (%)	Median	Range
Ethanol	9.0	26	7.0-112	12	380	153-780
Isopropanol	9.8	29	8.7-240	5	550	94-2,400
2-Butoxyethanol	1.2	2.6	0.7-17.8	4	62	14.7-990
Acetone	4.9	9.8	2.1-39	46	190	29-410
Formaldehyde	4.4	6.2	3.1-20	<15	40	20-174
Acetaldehyde	1.7	1.5	<0.1-3.2	<15	16.2	6.2-30
Hexanal	0.3	0.9	0.3-1.9	15	21	13.0-48
Isoprene	0.1	0.6	<0.1-2.6	22	10.9	5.5-28
d-Limonene	<0.1	0.8	0.2-4.9	4	27	17.0-79
D5 Siloxane[b]	<0.1	2.3	1.0-7.4	6	230	114-300
ΔCO_2 (ppm, $mg\ m^{-2}\ h^{-1}$)		210	122-510	--	56	35-80

[a]Summary for 21 samples collected near steady-state conditions. [b]D5 = defined in text.

The first four rotated principal components (PCs) of the analysis of VOC and CO_2 emission factors accounted for 79% of the total variance (Table 2). Tentative source identifications were made based on the most highly correlated compounds for each vector and information on likely sources. PC1 with high loadings of acetone, isopropanol and 2-butoxyethanol most likely represents the emissions of solvents from cleaning products (Zhu et al., 2001). Formaldehyde also has a high loading on PC1. PC3 with high loadings of

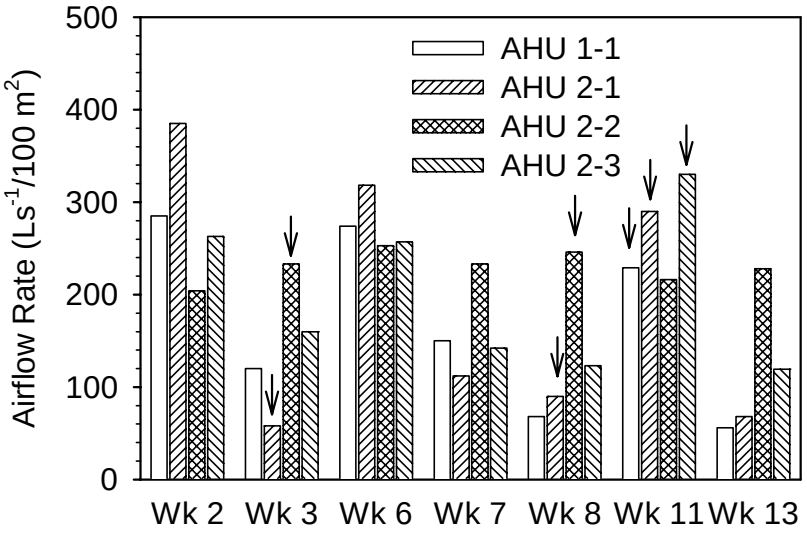

Figure 1. Normalized airflow rates in four AHUs on 7 sampling days. Arrows indicate non-steady state conditions.

decamethylcyclopentasiloxane (D5) and CO_2 presumably represents occupant sources as CO_2 is from breath and D5 is used in personal deodorants and has been associated with occupancy (Shields et al., 1996). Formaldehyde also is correlated with PC3. Isoprene is not correlated with this vector, although isoprene is a predominant VOC in breath (Fenske and Paulson, 1999), and isoprene concentrations were generally consistent with an occupant source. d-Limonene, which is highly loaded on PC4, is used as an air freshener and as an odorant and active ingredient in cleaning products (Zhu et al., 2001). Ethanol and formaldehyde are negatively correlated with this vector. The source of PC2 could not be deduced. Acetaldehyde and hexanal are both emitted by composite wood products (Hodgson et al., In press). However, wood products typically emit hexanal at much higher rates than acetaldehyde rather than the similar rates seen here. Also, wood products are sources of formaldehyde, which is negatively correlated with this vector. Ethanol and isoprene are both emitted in breath, but the ratio of their emission rates does not match this source (Fenske and Paulson, 1999).

The results of the PC analysis suggest that indoor air chemistry related to ozone may have impacted the concentrations of some VOCs. On the 7 sampling days, maximum 8-h values of outdoor ozone, obtained from a regional monitoring station, ranged between 25 and 42 ppb. Indoor ozone levels vary with ventilation rate and are frequently 20-70% of outdoor levels due to surface removal and homogeneous reactions with other chemicals in air (Weschler, 2000). At 50 ppb, the half lives of isoprene and d-limonene when reacted with ozone are 13 and 0.75 h, respectively (ibid.). Formaldehyde is a major product of these reactions. This is one explanation for the negative correlation of formaldehyde with d-limonene and isoprene in PC4. It is also a possible explanation for the week relationship of isoprene with CO_2 in PC3.

Table 2. Loadings of the first four rotated principal components for an analysis of emission factors of abundant VOCs and CO_2.

Compound	PC1	PC2	PC3	PC4
Ethanol	0.21	0.72	0.26	-0.25
Isopropanol	0.92	0.076	0.031	0.117
2-Butoxyethanol	0.90	-0.169	0.25	-0.078
Acetone	0.88	0.32	0.012	0.163
Formaldehyde	0.56	-0.40	0.62	-0.27
Acetaldehyde	-0.051	0.84	-0.177	-0.027
Hexanal	-0.007	0.84	0.147	0.100
Isoprene	0.081	0.54	0.064	0.43
d-Limonene	0.100	-0.073	0.24	0.88
D5 Siloxane	-0.056	0.081	0.84	0.24
CO_2 (ppm)	0.28	0.26	0.81	0.143
Variance (%)	32	23	14	10
Cum. Var. (%)	32	55	69	79

The In-Out concentrations of six VOCs variously identified with the range of source categories are plotted in Figure 2 versus normalized airflow rate. Only steady state data is included. The concentrations of isopropanol and 2-butoxyethanol, two compounds with likely cleaning product sources, showed little relationship with ventilation; although, the two highest isopropanol values occurred at the lowest airflow rates. Formaldehyde concentrations also were not apparently associated with ventilation as the concentrations in the zones served by AHUs 2-2 and 2-3 were nearly identical and constant throughout the study. Formaldehyde concentrations in the zones served by AHUs 1-1 and 2-1 were mostly higher possibly indicating a localized source; but, they were not associated with ventilation. Concentrations of acetaldehyde, hexanal, ethanol and isoprene (last two not shown) in zones served by AHUs 1-1, 2-1 and 2-3 exhibited trends of higher concentrations at lower flow rates to

varying degrees. The concentrations of d-limonene were not strongly associated with ventilation.

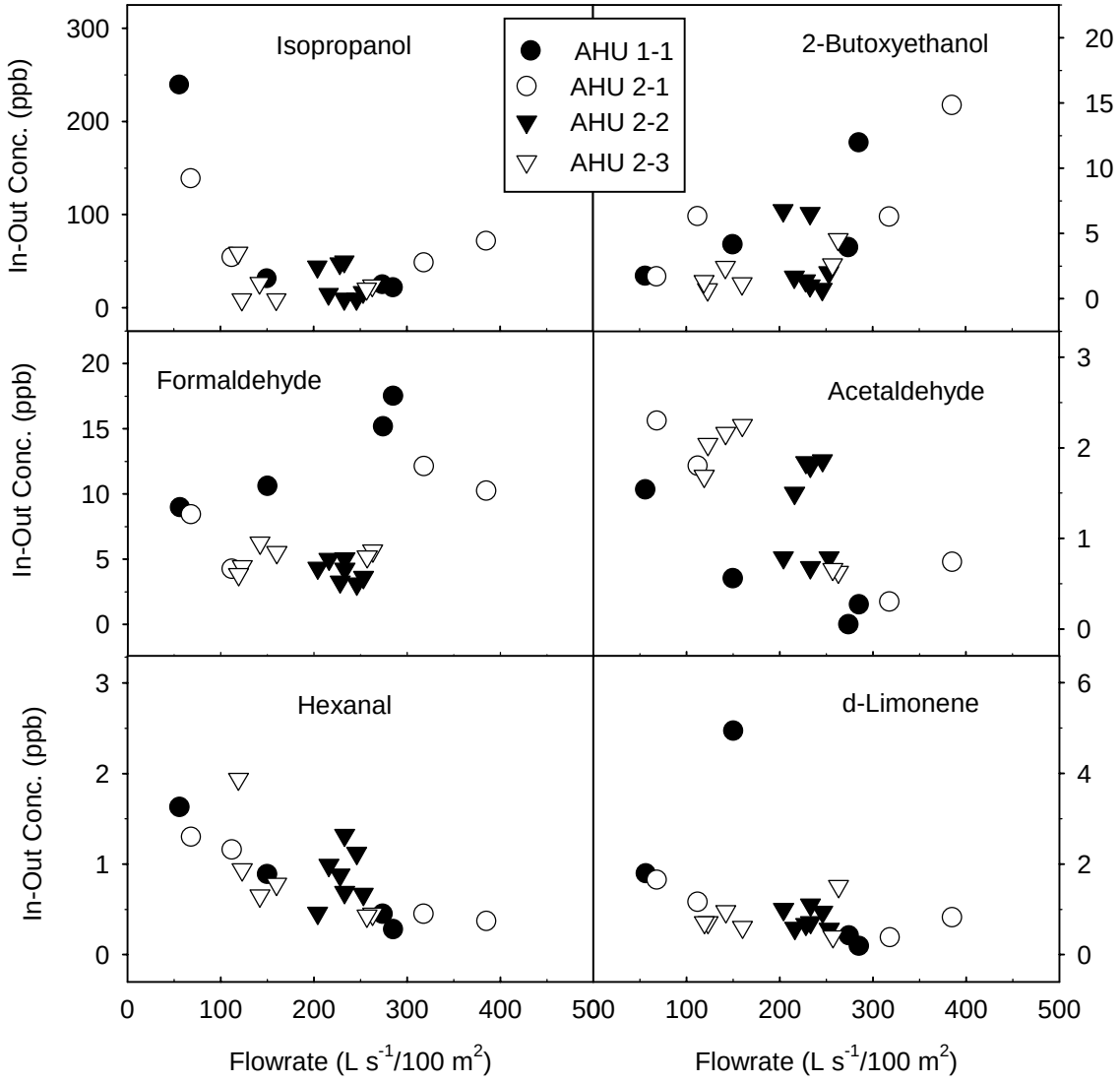

Figure 2. Indoor minus outdoor VOC concentrations in four AHUs on 7 sampling days versus normalized AHU airflow rates.

Understanding sources and evaluating the efficacy of ventilation for controlling the concentrations of VOCs emitted by these sources is difficult, particularly for large buildings. Direct relationships between VOC concentrations and ventilation may be obscured by a number of factors in addition to homogeneous air chemistry. There may be overriding temporal and spatial variations in source strengths. For example, concentrations of compounds that are solvent constituents of products used intermittently or sporadically and occasionally in large quantity would not be expected to exhibit strong inverse relationships with ventilation. This was apparently the case for acetone, isopropanol and 2-butoxyethanol. Ventilation and local airflow rates may influence VOC emissions from wet products applied to surfaces and from building materials, furnishings and other solid sources. The sorption of

VOCs onto surfaces in the building and their later release when bulk air concentrations decline also directly links VOC emissions with ventilation. For some common VOC/material combinations, this effect is predicted to be relatively large (Zhao et al., In press). These processes, plus imperfect air mixing and air chemistry, reduce the effectiveness of ventilation for controlling VOC sources.

CONCLUSIONS

Differences in the concentrations of most of the abundant VOCs in the building on 7 days of sampling could not be directly predicted by differences in ventilation rates on those days. Temporal and spatial variations in cleaning products likely obscured the relationship between concentration and ventilation for solvents. For other compounds, it is likely that effective emission rates increased with ventilation due to their re-emission from sinks. Homogeneous chemistry also may have altered the relationship between ventilation and concentrations of some reactive and product compounds. These results emphasize the importance of source control for limiting the concentrations of VOCs in buildings. Control procedures include use of low emitting materials to finish and furnish interiors, use of low-emitting cleaning products, and avoidance of products containing highly reactive chemicals.

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LONG TERM PERFORMANCE OF RADON MITIGATION SYSTEMS

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ABSTRACT
Researchers installed radon mitigation systems in 12 houses in Spokane, Washington and Coeur d'Alene, Idaho during the heating season 1985 –1986 and continued to monitor indoor radon quarterly and annually for ten years. The mitigation systems included active sub-slab ventilation, basement over-pressurization, and crawlspace isolation and ventilation. The occupants reported various operational problems with these early mitigation systems. The long-term radon measurements were essential to track the effectiveness of the mitigation systems over time. All 12 homes were visited during the second year of the study, while a second set 5 homes was visited during the fifth year to determine the cause(s) of increased radon in the homes. During these visits, the mitigation systems were inspected and measurements of system performance were made. Maintenance and modifications were performed to improve system performance in these homes.

INDEX TERMS
Radon mitigation, Sub-Slab Ventilation, Crawl Space Ventilation, Basement Over-pressurization, Long-term Radon Measurement

INTRODUCTION
A variety of approaches (described subsequently) have been used to reduce indoor radon concentrations, generally by reducing the rate of radon transport into the house from the surrounding soil. Much prior research has been described on the initial performance of these mitigation technologies, but only a few studies of long-term performance have been completed (Nitschke, et al.1988), (Scott and Robertson,1991), (Prill et al.1990). In 1985-86, we installed active mitigation systems in 12 houses (Turk et al. 1991). This paper reports the findings of a 10-year study of the performance of these systems. The resulting information will contribute to improvements in system design and installation practices.

METHODS
The follow-up radon measurements were made using alpha track detectors mailed to the occupants with detector placement and removal instructions, and data/comment cards for occupants to provide feedback on system noise, vibration, comfort issues, maintenance performed, occupant adjustment of fan switches and speed controls, crawlspace vents open or closed, and house remodeling.

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Quarterly and annual radon measurement results were immediately provided to the occupants of the homes. Occupants of those houses with indoor radon concentrations exceeding 148 Bq/m^3 were advised to take action to reduce the concentration to below 148 Bq/m^3, and were offered guidance and technical assistance.

Active Sub-slab Ventilation Systems: Seven houses were mitigated with active sub-slab ventilation systems. One house (ESP119) was mitigated with a pipe system installed from the exterior, routed under the footing and terminating under the basement slab floor. This system was operated in the conventional depressurization mode (Active Sub-Slab Depressurization or ASD). The other six houses were operated in pressurization mode (ASP) with the mitigation system fan mounted in reverse, creating a positive pressure in the pipe(s), forcing outside air beneath the slab floor. This technique was adopted in these six houses only after it was discovered that sub-slab depressurization systems were less effective in the highly air-permeable soils found in the Spokane and Rathdrum Prairie areas. ASP pipes were installed through the basement concrete slab floor into a pit filled completely with pea gravel. Screens at the outdoor intake were used to prevent, birds, rodents, insects, and debris from entering the pipes. In addition to pipes installed through the basement slab floor, house ESP111 had a second ASP system installed exterior to the house, with the pipes routed under the footing and terminating under the basement slab floor. The two ASP systems in house ESP120 were fitted with pleated filters at the pipe inlets to reduce particulate matter build-up in the pipes and on the gravel interface at the pipe outlet under the concrete slab floor.

Basement over-pressurization: Five houses were modified in the original experimental phase of the project to allow the basement zone to be pressurized relative to the surrounding soil. Four of these houses had central forced-air heating systems. An auxiliary axial fan was added to these duct systems to force main level air into the basement. These systems reduced radon concentrations in these homes at or below the target of 148 Bq/m^3 at the end of the experimental phase of the study. The fifth house, ECD027, has a dirt floor cellar used as storage. Original mitigation included adding insulation and a well-sealed membrane on the ceiling of this cellar. An axial fan was installed to create a positive pressure of 5 to 9 Pa in this zone relative to the outside using unconditioned outside air.

Crawl space Ventilation: Five of the homes were built with partial crawl spaces. During the experimental phase of the study, all crawl spaces were vented and a polyethylene membrane carefully installed on the surface of the crawl space soil as a soil gas retarder.

RESULTS

Increasing radon concentrations and system problems reported by the occupants prompted researchers to perform relatively minor modifications and maintenance to some of the systems two years after the initial experimental study was completed (Prill et al, 1990). In the basement pressurization houses, clothes dryer vent flaps were either missing, stuck open, or were being forced open by the positive pressure being created in the basements. To prevent loss of pressure, carefully weighted flaps were added to these vents. The flow rate of the basement pressurization fans was found to have decreased 20 to 25 percent in this set of

homes in spite of the fact that the measured effective leakage area of these basements was unchanged. Fan speeds had been reduced or systems turned off for various reasons. Occupants in house ESP116 reported occasional back-drafting of the airtight-rated wood stove located on the main level. Providing additional combustion make-up air to the main level of the home solved the woodstove back-drafting. Mitigation resulted in radon reductions from the initial baseline of 1,665 down to 22.2 Bq/m^3.

The year-two modifications in the active sub-slab ventilation houses included minor adjustments to fan installation details to correct noise and/or vibrations (House ESP111 ASP system was turned off due to fan vibration). Filters on ASP systems had became loaded with debris resulting in decreased system air velocity and pressure. Sealants at the ASP pipe penetrations had failed resulting in air leaks and reduced system pressures. These joints were cleaned and resealed with a superior caulking product. Radon concentrations in all six of the ASP homes had exceeded the target concentration of 148 Bq/m^3 in at least one measurement period. Radon measurement results are presented in Tables 1 and 2. In houses ESP108 and ESP113 easily accessible ASP pipes were severed near the slab and a visual inspection was made. Debris carried by the outside air had built up on the gravel layer at the pipe terminus. After debris removal there was a significant increase in air velocities and a decrease in system pressures (Prill, et al.1990). It is presumed that the decrease in system performance in the other four ASP houses was a result of similar debris build-up.

A second set of modifications in the five homes with ASP systems were performed during the fifth year (1991) of the follow-up study prompted by radon concentrations in all of the ASP homes exceeding the target concentration of 148 Bq/m^3 during at least one measurement period. The inspections revealed that once again pressures in the systems had increased, and air velocities decreased, resulting from the build-up of debris at the terminus of the ASP pipes at the sub-slab gravel layer.

Clearly, removal of debris from the systems was not adequate to maintain system effectiveness, nor was frequent cleaning at the pipe/soil interface practical. Debris was removed and 22 to 44 liter pits were created in the ASP systems in five houses. Significant increases in pressure field extension by adding a 22 to 44 liter pit at the pipe terminus of active sub-slab ventilation systems in existing houses has been documented. (Prill, et al.1992). The ASP fan speeds were recorded and the pressure and air velocity were measured before and after modifications. House ESP101 ASP pressures decreased by a factor of 3 and air velocity increased by a factor of 5.5 after pits were created at two of the 3 pipes. In house ESP108 ASP pressures decreased by a factor of 1.4 and air velocity increased by a factor 43.6 after pits were created at three of the four pipes. House ESP111 ASP pressures decreased by a factor of 2.6 and air velocity increased by a factor of 5.7 after pits were created in the interior system (exterior system not accessible). House ESP113 ASP pressures decreased by a factor of 5.5 and air velocity increased by a factor of 7.2 after pits were created. House ESP120 ASP pressures decreased by a factor of 4 and air velocity increased by a factor of 21 after pits were created in one of the two systems. ASP house ECD026 was not visited (or modified) the fifth year.

Table 1. Radon concentrations (Bq/m3) in homes with active sub-slab ventilation

House ID	ESP 101	ESP 108	ESP 111	ESP 113	ESP 119	ESP 120	ECD026
Location	B, M	B, M	B, M	B, M	CS, B, M	B, M	CS, B, M
Winter-Post Mit. 85-86	74, 37	37, 37	111, 74	ND, ND	ND, 37, ND	111, 74	ND, 111, 111
Annual 4/87-4/88	629, 407	407, 296	555, 555	296, 222	37, 180, 111	740, 555	111, 111, 111
Annual 9/88 - 10/89	148, 111	148, 148	ND, ND	148, 111	37, 74, 37	ND, ND	ND, ND, ND
Annual 4/90 - 4/91	666, 333	296, 222	ND, ND	296, 296	37, 111, 37	888, 666	148, 148, 148
Annual 4/91 - 4/92	222, 111	111, 111	222, 148	37, 37	37, 111, 74	296, 185	111, 148, 148
Annual 4/92 - 4/93	111, 37	333, 185	407, 407	74, 74	37, 110, 37	111, 74	148, 185, 148
Annual 4/93 -4/94	111, 74	111, 74	925, 740	74, 74	37, 74, 74	185, 111	185, 222, 222
Annual 94 -95	148, 111	74, 74	925, 666	74, 74	37, 111, 74	185, 148	296, 296, 296
Annual 6/95 - 6/96	259,ND	74, 37	740, 703	74, ND	ND, 37, 111	666, 444	ND, 185, 185

KEY: ND: No Data Location: CS = Crawl Space; B = Basement; M = Main Level

Table 2. Radon concentrations (Bq/m^3) in homes with basement over-pressurization.

House ID	ESP 116	ECD 027	ECD 153	NCD 077	NSP 204
Location:	B, M	M	CS, B, M	CS, B, M	CS, B, M
Winter-Post Mit.85-86	ND, 74	37	ND, ND, 37	ND, ND, 74	ND, ND, 111
Annual 4/87-4/88	777, 180	180	150, 300, 260	180, 440, 220	370, 330, 180
Annual 9/88 - 10/89	ND, ND	37	ND, ND, ND	148, 259, 111	296, 296, 148
Annual 4/90 - 4/91	ND, ND	296	185, 74, 74	222, 481, 222	629, 407, 333
Annual 4/91 - 4/92	ND, ND	37	222, 111, 74	148, 592, 296	518, 407, 333
Annual 4/92- 4/93	777, 515	185	259, 198, 148	148, 518, 259	777, 629, 407
Annual 4/93 -4/94	407, 962	148	333, 185, 222	259, 444, 222	851, 703, 481
Annual 94-95	ND, ND	222	296, 148, 148	148, 444, 259	740, 592, 407
Annual 6/95 - 6/96	ND, ND	2146	296, 259, 222	ND, ND, ND	777, 592, 407

KEY: ND: No Data Location: CS = Crawl Space; B =Basement; M = Main Level

CONCLUSION AND IMPLICATIONS

The mitigation systems in this set of homes were designed and installed during the early years of radon mitigation and were experimental. It is important to note that during the experimental phase of this study the fan speeds and pressures of the mitigation systems were adjusted to control the indoor radon at just under the target of 148 Bq/m^3. These systems were thus intentionally not over-sized nor particularly robust, and hence more likely to be sensitive to occupant adjustment of fan speeds, lack of maintenance, decreased fan performance over time, and other factors. However, most mitigation strategies require some level of maintenance, and fan-powered active systems experience decreased fan performance over time and eventually fan failure.

The occupants of these homes contributed to the reduced performance of the mitigation systems by actions including: turning fans off, reducing fan speeds, defeating pressurization regimes, closing crawl-space vents, and remodeling activities. When notified of increasing radon concentrations most of the homeowners with ASD systems responded promptly to improve the performance of the mitigation systems. However, the homeowners with the basement pressurization systems did not respond, nor did the homeowner in house ESP111, in spite of offers of technical assistance from the researchers.

Active Sub-Slab Ventilation: After the second visit (year five) when pits were created at the ASP pipe terminus, houses ESP108 and ESP113 have remained below the target of 148 Bq/m3 annual average. System fans failed in houses ESP101 and ESP120 resulting in basement level annual average radon concentrations increasing from 111 Bq/m^3 to 259 Bq/m^3 and 111 Bq/m^3 to 666 Bq/m^3 respectively (fans were eventually replaced). Radon concentrations continued to increase over time in house ESP111 where the exterior ASP system was not accessible for modification.

Active sub-slab pressurization systems can be more effective than ASD systems in some buildings where construction details and/or highly permeable soil conditions exist, however this study suggests that ASP system effectiveness can be significantly reduced by the accumulation of debris deposited on the sub-slab soil/gravel. Properly designed low-flow ASP systems promise to offer highly effective and energy efficient radon control (Fisk et al.1995).

The one house in this study (ESP119) with active sub-slab depressurization (ASD) continued to remain below the target of 148 Bq/m^3 annual average for the entire duration of the study, except for one measurement. The system performance was not measured nor were modifications made, and according to data/comment cards returned by the occupant, the system fan has not been repaired nor replaced.

Basement Over-Pressurization: The homeowners have not reported any evidence or concerns in terms of structural problems associated with the basement pressurization systems. These systems, while very effective at reducing radon entry when designed, installed, and operated properly are easily defeated by occupants leaving basement doors open, not maintaining door seals at the basement door, turning the fan off, or changing the

balance of the supply and return systems. Three of these five homes performed poorly in terms of maintaining radon concentrations below the target of 148 Bq/m^3. Measurements of fan performance after two years found decreases in air flow rates of 20 to 25 %. Initial reports of back-drafting from the main level wood stove in house ESP116 required adding additional combustion air to that zone. Therefore the reliability and practicality of these systems must be questioned. Two of the homes, ESP116 and NCD 077, were sold during the study. The new owners/occupants were not informed by the sellers of the radon system, or follow-up study. The new occupants (year 1988) of house ESP116 were uninterested in the study and did not reliably deploy detectors, or report system and/or house operation. Subsequent radon concentrations in house ESP116 suggest that the system was either not operated or the pressure regime was defeated for reasons that are not clear. The radon concentrations in house ECD027 remained below the target of 148 Bq/m^3 except during those periods where the fan was off due to cold weather and during an extensive remodel of the home in 1995 and 1996.

With thousands of radon mitigations systems installed in homes across the U.S. over the last two decades, the findings from this small set of homes suggest additional long-term radon follow-up studies are warranted.

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METHOD FOR MEASURING THE SIZE DISTRIBUTION OF AIRBORNE RHINOVIRUS

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ABSTRACT
About 50% of viral-induced respiratory illnesses are caused by the human rhinovirus (HRV). Measurements of the concentrations and sizes of bioaerosols are critical for research on building characteristics, aerosol transport, and mitigation measures. We developed a quantitative reverse transcription-coupled polymerase chain reaction (RT-PCR) assay for HRV and verified that this assay detects HRV in nasal lavage samples. A quantitation standard was used to determine a detection limit of 5 fg of HRV RNA with a linear range over 1000-fold. To measure the size distribution of HRV aerosols, volunteers with a head cold spent two hours in a ventilated research chamber. Airborne particles from the chamber were collected using an Andersen Six-Stage Cascade Impactor. Each stage of the impactor was analyzed by quantitative RT-PCR for HRV. For the first two volunteers with confirmed HRV infection, but with mild symptoms, we were unable to detect HRV on any stage of the impactor.

INDEX TERMS
Bioaerosols, Infectious disease, Measurement methods, Quantitative RT-PCR, Rhinovirus

INTRODUCTION
Indoor airborne bioaerosols contribute to a broad range of health effects including communicable respiratory illness. The majority of the US population has one or more respiratory illnesses annually (Turner, 1997). The important bioaerosols include human-produced droplet nuclei containing infectious virus and bacteria, non-infectious bacteria and fungi (e.g., molds), and allergens from plants, dust mites, insects (e.g., cockroach), and animals (e.g., pets). Measurements of the airborne concentrations of these bioaerosols are critical for research on the relationship of exposures to health effects, for studies of the influence of building characteristics on exposures, and for evaluations of exposure mitigation measures. Only very limited data exist regarding the size distribution of human-produced droplet nuclei and no data are available on droplet nuclei containing human rhinovirus (HRV).

There are enormous limitations in the sampling and analysis methods used traditionally for many of these bioaerosols. Airborne virus measurement has required very large samples to overcome sensitivity limitations and culturing within the tissue or cells of a host (e.g. human tissue); hence, these measurements are extremely rare. Polymerase chain reaction (PCR) is extremely sensitive, and the use of primers with DNA sequences unique to the target of interest make it also very specific. Recent development of a real-time PCR system makes quantitation of the target sequence faster and easier. The use of PCR for applied health research began primarily with the analysis of body fluids or nasal washings to diagnose disease. For example, Johnston *et al*. (1995) used PCR to detect common respiratory viruses,

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including rhinovirus, in nasal aspirates and found that asthma exacerbation in children (9-11 years) was associated with a respiratory infection about 80% of the time. PCR has also been used recently (Aintablian *et al*., 1998, Sawyer *et al.*, 1994, Kashima *et al.*, 1991, and Sawyer *et al.*, 1995) for the non-quantitative or semi-quantitative detection of selected viruses and bacteria in air samples (e.g., Varicella-zoster virus, Respiratory Syncytial virus, M. Tuberculosis bacteria).

The goals of this study are to develop a quantitative reverse transcription-coupled PCR (RT-PCR) assay for human rhinovirus (HRV), to detect airborne HRV in bioaerosols, and to use this assay to learn about the aerodynamic size distribution of airborne particles with rhinovirus. Most of these particles are expected to be droplet nuclei containing rhinovirus that are produced by infected people during coughing and sneezing; however, some of these droplet nuclei may attach to other airborne particles. Dick *et al*. (1987) demonstrated that rhinovirus infections are transmitted, in part, through inhalation of virus. A review of ten epidemiologic studies found that building characteristics, such as ventilation rates, are significantly associated with the prevalences of respiratory illnesses experienced by building occupants, with most adjusted relative risks or odds ratios between 1.3 and 2.0 (Fisk 2000). The principal explanation for these findings is that building characteristics influence exposures to droplet nuclei with viable virus. The aerodynamic size of these droplet nuclei determine their range of transport indoors, rates of loss from indoor air via deposition, location of deposition in the respiratory system, and the efficacy of control measures such as filtration. Very few data however, are published on the size distribution of human-produced droplet nuclei (Brosseau *et al*., 1994).

METHODS

Quantitative RT-PCR Assay: Precautions were taken to minimize contamination during sampling, RNA extraction, and reaction mix preparation (Kwok and Higuchi, 1989). A negative control was included in each set of samples processed. Before reverse transcription, RNA secondary structure was removed by heating the sample RNA with the forward primer HRV01F (Table 1) for 4 minutes at 72°C, then immediately chilling the sample on an ice bath. The RNA/primer mixture was reverse transcribed using 200 units of Moloney-murine leukemia virus reverse transcriptase (M-MLV RT) and 30 units of a ribonuclease inhibitor for 1 hour at 40°C. The resulting cDNA was stored at -80°C or amplified immediately.

Table 1. HRV RT-PCR Primer sequences.

Primer	*Sequence*	*Position*
HRV01F	5'-GAAACACGGACACCCAAAGTAG	531-552
HRV01D	5'-TAGCCTGCGTGGCTGCC	349-366

The quantitation of the 203 base pair (bp) HRV PCR product generated by primers listed in Table 1 was performed using a rapid air microcapillary cycler with real-time product detection by fluorescence (LightCycler, Roche Molecular Biochemicals). cDNA was amplified using the LightCycler-FastStart DNA Master Mix reagent (Roche Molecular Biochemicals) with a final concentration of 12.5 pmol each primer and 3 mM $MgCl_2$. Thermal cycling conditions are given in Table 2. A serial dilution of the external quantitation standard (HRV QS) is included with each group of samples that are amplified.

Table 2. Thermal Cycling Conditions for Quantitative HRV RT-PCR Assay.

Segment	*Step*	*Conditions*		*Cycles*
		+Temp.°C	*Time (sec.)*	
denaturation	1	95	10 min.	1
initiation	1	95	10	4
	2	73	10	
quantitation	1	95	5	35
	2	69	15	
	3	72	10	
	4	84	5*	
melting	1	70	15**	1
+(0.1°C/sec.)	2	95	1	

+All temperature transition rates are 20°C/sec. unless indicated.
*single fluorescence reading
**continuous fluorescence acquisition

Quantitation Standard(QS): The HRV QS containing the 203 bp HRV target sequence was obtained by cloning a 425 bp fragment of the 5'untranslated region HRV genome into a bacterial cell line. Briefly, the 425 bp HRV fragment was ligated into a pGEM vector (obtained from Promega) creating a pHRV vector. Competent *E. coli* cells (JM109) were transformed with the pHRV vector. Using the proper selection media, the transformed bacteria were grown, colonies were selected and screened for the presence of the pHRV vector. Large quantities of the pHRV vector were isolated and purified. After linearizing the pHRV vector, the RNA HRV QS standard was produced by *invitro* translation. A fluorescent assay was used to accurately determine the concentration of the HRV QS.

Bioaerosol Sampling: Adult volunteers exhibiting symptoms of a common head cold were recruited from employees according to Human Use Protocols. Volunteers spent 2 hours in a $27m^3$ research chamber while aerosol samples were collected. The research chamber environment had an average temperature of 23°C and a relative humidity of 35% and was supplied with outside air. A small floor fan was set at low speed to provide mixing, while the sampler was placed on a 3 ft. high computer table about 3 ft. from the subject. Chamber air was sampled with a six-stage viable cascade impactor (ThermoAndersen) with 50% size cuts of 7, 4.7, 3.3, 2.1, 1.1 and 0.65 microns. A 45 mm teflon-coated glass fiber filter (Gelman Pallflex, T60A20) was used as a final stage (to collect particles with sizes <0.65 microns). The impactor was prepared for sampling by exposing each side of every stage to a germicidal light for 30 minutes before use. A sterile glass petri dish containing 10 ml of sterile mineral oil was placed into each stage. A sampling rate of 28.3 lpm was maintained with a Gilian AirCon2 high volume pump and was calibrated with a dry gas meter (Rockwell International). A tape recorder was used to provide information on the periods of coughing and sneezing. After the sampling period, a nasal lavage was obtained from each nare of the volunteer. The lavage was stored at -20°C until analysis. The impactor and filter were stored at 4°C until analysis.

HRV RNA Isolation: RNA was isolated from lavage samples using Tri Reagent LS (Sigma Chemical Company) according to the manufacture's directions. The isolated RNA was resuspended in ribonuclease-free H_2O and then reverse transcribed immediately, as described

below. RNA was isolated from viable HRV type 89 (American Type Culture Collection) for use as a positive control. To recover HRV from each stage of the impactor, the petri dishes were rinsed 4 times with 3 ml chloroform each and transferred to a 50 ml polypropylene sterile centrifuge tube. The dishes were scraped each time with a sterile policeman to facilitate the transfer and resuspension of the particles. Each dish was washed one final time with 3 ml of RNA lysis buffer (Chomczynski and Sacchi, 1987). The two phases were mixed together by shaking and vortexing, then separated by centrifugation at 2,000g for 2 minutes at 15°C. The HRV RNA in the lysis buffer was recovered by alcohol precipitation as described by Chomczynski and Sacchi (1987). RNA was resuspended in ribonuclease-free H_2O and then immediately reverse transcribed.

RESULTS

The specificity of the HRV RT-PCR assay is determined by primer sequence. Primers HRV01F and HRF01D (see Table 1) were slightly modified from primer ncr2 and probe Rh of Andeweg *et al.*, (1999). The primers are located in regions of high genetic homology of the 5'untranslated region of the HRV genome as indicated by a genetic analysis and sequence alignment of 25 HRV strains (Andeweg *et al.*, 1999). As a result, this assay detects more than 90% of HRV strains and will not detect the genetically similar enterovirus family; however, several enterovirus-like HRV strains (87,14,37,72) are not detected. Computer-aided analysis of several different HRV strain sequences obtained from GenBank was used to confirm this observation.

Before beginning chamber studies, the HRV RT-PCR assay was tested on the nasal lavages of 6 volunteers with head cold symptoms and 1 volunteer with allergy symptoms. Four of the 6 head cold specimens were found to be HRV positive while the specimen from the allergy sufferer was negative and served as a negative control. Melting curve analysis at the end of the amplification serves to identify each product formed during PCR (see Figure 1). The melting curve is unique to each product and is dependent on length and nucleotide sequence of the DNA. RNA from HRV type 89 was used to confirm a positive lavage sample (5R) and the HRV QS standard. All have a melting temperature of 88°C. A negative sample (2R) shows non-specific product formation at lower temperatures.

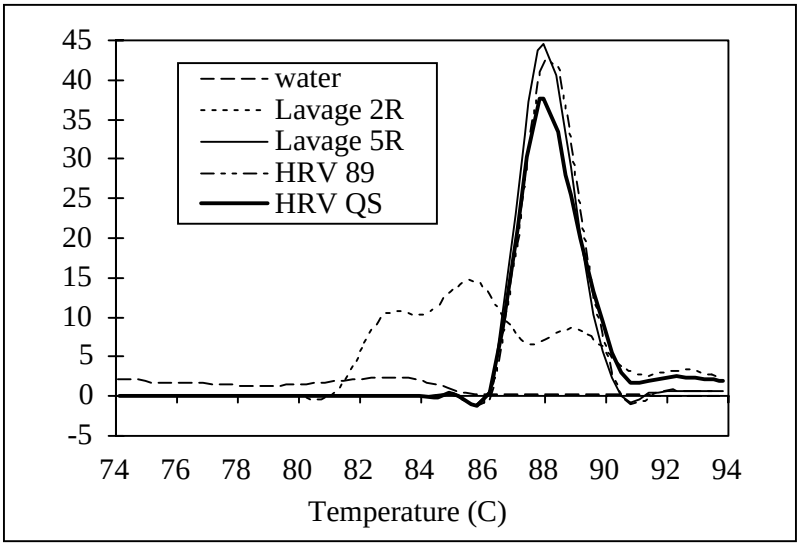

Figure 1. Melting Curve Analysis of HRV PCR Products.

During PCR there is an exponential accumulation of product; this phase of the reaction is used for quantitation. The concentrations of unknown samples are determined from the first cycle of the PCR run in which the signal can be distinguished from the background. By extrapolating this cycle number from a standard curve generated from a dilution series of the HRV QS (Figure 2), the concentration of an unknown sample may be determined. This remains true as long as the standard and unknown are amplified with the same efficiency as is true for this assay. A detection limit of 5 fg of HRV QS was observed. At this point non-specific product formation competes with HRV product accumulation and although detection is still possible at this level, quantitation is no longer valid. The standard curve is linear over a 1000-fold range and the assay has a variance of 8% at the 5000 fg level and 20% at the 50 fg level (n=3).

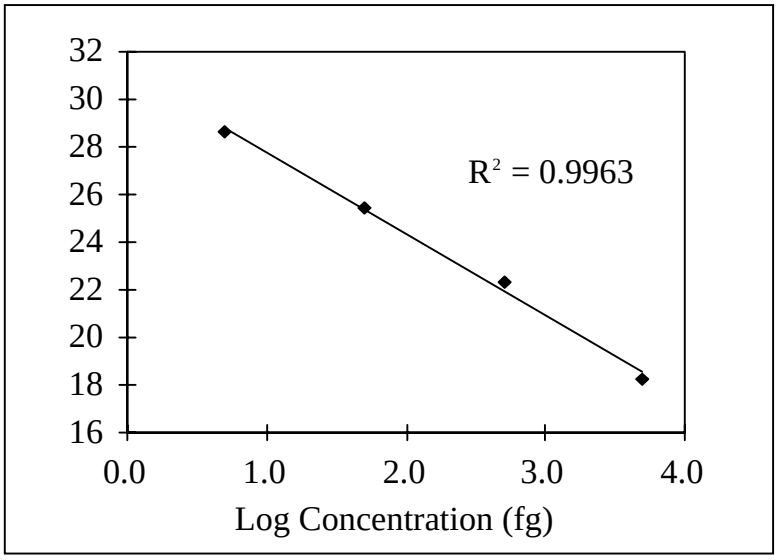

Figure 2. Standard Curve for Quantitation of HRV.

The HRV RNA quantitation standard was used to develop a sample extraction method. Filters and petri dishes were spiked with various amounts of the HRV QS and then subjected to 4 hours of sampling using the six-stage impactor. As a control, the HRV QS was isolated directly from the lysis buffer without any sampling or sample extraction. Recovery of 10 pg by the alcohol precipitation method from the impactor was 60% with a variance of 30% (n=3). In comparison, precipitation directly from the lysis buffer gave a recovery of 65%. Stages were extracted a second time to determine how much of the HRV QS remained. Only 1% of the initial amount spiked into the mineral oil was detected.

To date, we have recruited 4 volunteers exhibiting head cold symptoms to participate in the chamber experiment. Two of the volunteers displayed very strong cold symptoms, but were negative for HRV. The other two volunteers, who displayed only light symptoms (some coughing and no sneezing), were positive for HRV. However, no HRV could be detected on any stage of the impactor.

DISCUSSION

The most common bioaerosol measurement approach is the impaction of particles on culture media, followed by growth of these organisms. HRV identification by viral culture is difficult

and can take up to 3 weeks. Using a PCR based assay for the detection of HRV eliminates the need to recover viable organisms, increases sensitivity three-fold over viral culture systems, and reduces the assay time to two hours (Johnston *et.al.*, 1995). Existing HRV PCR assays are developed for clinical diagnostic purposes and are qualitative or semi-quantitative in nature. The HRV RT-PCR assay we describe here is validated as a strictly quantitative method with a detection limit of 5 fg and is capable of detecting over 90% of the more than 125 characterized HRV strains.

The HRV genome consists of 7,102 nucleotide bases, therefore 1 fg of HRV RNA corresponds to about 280 HRV organisms. Our calculations show that HRV viral production in the nasal mucosa of infected individuals is great enough to suggest the feasibility of detecting HRV in aerosol samples. We used the HRV QS to estimate the viral load in one nare of an individual to approximately $1x10^7$ HRV genome equivalents. Taking into account the assay sensitivity and sampling and recovery efficiencies, we need to collect $3x10^3$ HRV organisms or 0.03% of the viral load that an individual might produce.

One of the keys to successful analysis of HRV in aerosol samples is the improvement of the sample recovery process. Initial experiments with the HRV QS show that RNA is not degraded during the sampling process with the six-stage impactor; however, half of the RNA is lost during the alcohol precipitation step. We also examined RNA recovery from a commercially available solid-phase substrate, but recovery of the HRV QS at the 10 pg level was found to be only 10%. This method was not pursued. Currently, very little is known about viral production during an HRV-induced cold. Conflicting reports of virus shedding vary from 1 day before symptoms occur to between 2 days and 3 weeks after the onset of cold symptoms (Turner, 1997). One of our volunteers participated in the chamber experiment after experiencing cold symptoms for 4 days. Although this nasal lavage was weakly positive, it is unknown if viral shedding would have been greater at the onset of symptoms. It is also difficult to find volunteers that have not taken an antihistamine to suppress many of the cold symptoms that would normally produce an aerosol.

CONCLUSION

We developed a quantitative RT-PCR assay specific for HRV that can quantify the virus to 5 fg. We also can recover HRV RNA from previously spiked multistage impactors. This HRV RT-PCR assay is validated and is ready to be used in the detection of HRV in aerosols.

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RELATIONSHIP OF SBS-SYMPTOMS AND VENTILATION SYSTEM TYPE IN OFFICE BUILDINGS

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ABSTRACT
This paper provides a summary of current knowledge about the associations of ventilation system types in office buildings with sick building syndrome symptoms. Most studies completed to date indicate that relative to natural ventilation, air conditioning, with or without humidification, was consistently associated with a statistically significant increase in the prevalence of one or more SBS symptoms, by approximately 30% to 200%. In two of three analyses from a single study (assessments), symptom prevalences were also significantly higher in air-conditioned buildings than in buildings with simple mechanical ventilation and no humidification. The available data also suggest, with less consistency, an increase in risk of symptoms with simple mechanical ventilation relative to natural ventilation. The statistically significant associations of mechanical ventilation and air conditioning with SBS symptoms are much more frequent than expected from chance and also not likely to be a consequence of confounding by several potential personal, job, or building-related confounders. Multiple deficiencies in HVAC system design, construction, operation, or maintenance, including some of which cause pollutant emissions from HVAC systems, may contribute to the increases in symptom prevalences but other possible reasons remain unclear.

INDEX TERMS
HVAC, Sick building syndrome, Ventilation system type, Review article

INTRODUCTION
The primary objectives of this paper is to synthesize available literature on the associations of ventilation system types in office buildings with sick building syndrome symptoms and to evaluate potential explanations for the associations. This paper is based on the review by (Seppänen and Fisk, 2001).

In many studies, prevalence of sick building syndrome symptoms have been associated with characteristics of buildings and ventilation systems. One of the most important factors affecting indoor air quality is how the building is heated, ventilated and air-conditioned. In many cases, particularly in office buildings, these functions are integrated in one system. In this paper these systems are called HVAC systems (heating, ventilating and air-conditioning systems).

Studies of the associations of ventilation system types or features with health and perception outcomes have primarily been performed in buildings several years after construction, and risk factors could differ in new buildings. Most studies have been cross-sectional, with data on health (or perception) outcomes, ventilation system characteristics, and other relevant factors collected in multiple buildings and these data analyzed statistically to determine the

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strength and uncertainty in the associations of health outcomes with the type of HVAC system. A weakness of this study design is that many factors other than ventilation system type vary among the buildings and may influence the health outcomes, confounding the association of HVAC system type with the health outcome. The better cross-sectional studies control for many potential confounding factors in the study design or data analyses. Unfortunately, some studies have controlled for few or no confounding factors in statistical analyses. Another inherent weakness of cross-sectional studies is that occupants with substantial adverse health effects from exposures in a building may more frequently be absent or quit working in the building. For these reasons, cross sectional studies can find statistical associations but, without other supporting findings, such studies cannot confirm causal relationships.

APPROACH

General approach

The overall approach was to identify relevant papers for review, to set criteria for studies to be included, to analyze the available information from studies meeting the inclusion criteria and process the results into a common format, and finally to draw conclusions.

Study inclusion criteria

The review included only studies of office buildings, although some information is available from schools, residences, etc. Most of the studies have used SBS symptoms as the outcome, and we excluded other outcomes from the review. We also excluded studies that did not perform a statistical test to determine if there were statistically significant differences in symptoms between occupants of buildings with different HVAC types.

The power of a cross-sectional study increases with the number of study buildings or study spaces with different HVAC systems, and also with the number of occupants included in the study. Increased power reduces effects of random error, but does not reduce systematic bias. For this review, we excluded from consideration any cross-sectional study with less than two buildings in any included HVAC-type category. We also excluded studies primarily containing complaint buildings, because we suspected that the widespread concerns about health in complaint buildings could decrease the validity of self-reported symptoms.

Some published experimental studies involved movement of subjects from building to building or with replacement of HVAC systems, with analyses of the changes in symptom prevalences within subjects. Some potential confounding is eliminated by within-subject analyses; for example by personal and job-related factors, which are unchanged during the experiment. However, there is still a possibility of confounding by many parameters which may have varied among the experimental periods, such as building characteristics, indoor temperature, outdoor conditions or job stress. We considered these sources of potential bias so significant that we excluded from our review any studies with movement of the study population between buildings or with replacement of HVAC systems. Another weakness of these studies is that occupants' awareness of the HVAC and environmental changes may have influenced their symptom reporting on questionnaires.

All studies that fulfilled the criteria described above were included in our review whether or not statistically significant associations were reported.

RESULTS

Tables 1 and 2 summarize the major features and findings of studies included in this review. A study may have performed multiple analyses (called assessments) between different groups of HVAC types or analyzed different subsets of study data (e.g. a natural ventilation group was compared with air conditioning group and also with a simple mechanical group). Each assessment is presented on an individual row in Table 1 or 2.

Association of HVAC system types with SBS symptoms

Table 1 presents the assessments comparing symptoms among occupants of air-conditioned buildings with those in naturally-ventilated or simple-mechanically-ventilated buildings. Table 2 presents the assessments comparing symptoms associated with simple mechanical ventilation to symptoms associated with natural ventilation. Tables 1 and 2 provide the following data: a) the number of symptoms or symptom groups in the analyses; b) the number of symptoms that were statistically significantly associated with HVAC system type; and c) when available, the range of relative risks or odds ratios for statistically-significant associations. Additionally, the presence or absence of statistically-significant associations of HVAC system types with outcomes is illustrated graphically within the tables using an adaptation of the format of (Mendell, 1993). HVAC system types are indicated by circles located in the appropriate columns. When the type of humidification was uncertain or included multiple types, the circle was replaced with a horizontal bar extending across the applicable columns. Within these tables, shading of a circle or horizontal bar (relative to no shading), indicates that the study found a statistically-significant increase in prevalence of one or more symptoms among occupants with that HVAC system type relative to buildings with the reference-type of HVAC. Unshaded circles at both ends of a connecting line indicate that the subjects served by different types of HVAC systems did not have significantly different symptom prevalences. The numbers adjacent to the circles denote the number of buildings in the assessment with that type of HVAC system. Blank spaces in the tables indicate that the information was not reported.

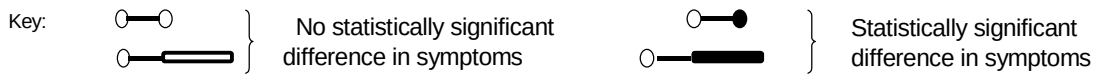
Referring to Table 1, 16 of 17 assessments found a statistically significant increase in the prevalence of one or more symptoms with air conditioning relative to natural ventilation. Nine of these assessments controlled for two or more types of confounding factors, and eight of the nine found a significant increase in symptoms with air conditioning. Two of three assessments found a statistically significant increase in the prevalence of symptoms with air conditioning relative to simple mechanical ventilation without air conditioning; however, no significant increase in symptom prevalences was found in the assessment with the largest number of buildings. Air conditioning with or without humidification was associated with significant increases in symptom prevalences. The studies provided minimal information to assess the hypothesized increase in risks with various types of humidification. In 12 of 20 assessments, air conditioning was associated with a significant increase in the prevalence of a majority of the symptoms or symptom groups. Most of the relative risks or odds ratios were between 1.3 and 3.0, indicating roughly up to 30% to 200% increases in symptom prevalences in the air conditioned buildings.

The results of the nine assessments that did not involve air conditioned buildings are provided in Table 2. In five of seven assessments that compared simple mechanical ventilation to natural ventilation or to sets of buildings with both natural and exhaust ventilation, prevalences of one or more symptoms were statistically-significantly higher with simple mechanical ventilation. The study with the largest number of buildings (Sundell et al., 1994) did not find a significantly higher symptom prevalence with simple mechanical ventilation;

Table 1. Comparison of SBS symptom prevalences with and without air conditioning.

Reference		Study and Building Characteristics					Ventilation System Type								Results	
								Mechanical Without AC			Air Conditioning					
First Author	Year	Controlled confounders#	No of respondents in comparison	Sealed or openable windows*	Smoking	Recirculation *	Natural Ventilation	Mechanical exhaust	Simple mechanical, no humidification	Simple mechanical with humidification	No Humidification	Steam Hum.**	Evaporative Hum.	Spray Hum.	Number of Symptoms with signicantly higher prevalences in assessment^^	Range of risk ratio or (odds ratio) for outcomes
Jaakkola	95	P,W,B	868	O		Y/N	7				9				2 of 14 S	1.5-2.6
Mendell	96	P,W	710	S	N	Y	3				6				6 of 7 S	1.6-5.4
Burge^	87	none	1459	S/O			11				10				10 of 10 S	(1.3-2.1)
Harrison^	87	none	1044	S		Y/N	8				6				6 of 6 S	(1.7-2.9)
Zweers	92	P,W,B	2806	S/O	Y		21								5 gr. of S	1.5-1.7
Jaakkola	95	P,W,B	335	O		Y	7					2			3 of 14 S	(1.9-2.5)
Burge^	87	none	863	S/O			11					4			8 of 10 S	(1.3-2.1)
Zweers	92	P,W,B	3573	S/O	Y		21								5 of 5 gr. of S	1.3-1.9
Jaakkola	95	P,W,B	559	O		Y/N	7						3		3 of 14 S	(2.0-2.7)
Teeuw	94	none	927	S/O		Y/N	7						7		5 of 8 S	1.4-2
Burge^	87	none	1991	S/O			11							15	10 of 10 S	(1.4-2.2)
Finnegan^	87	none	787	S	Y	Y/N	3							3	6 of 11 S	(2.5-4.8)
Harrison^	87	none	2080	S		Y/N	8							13	5 of 6 S	(2.1-3.2)
Hedge^	84	none	1214				2							2	2 of 2 S	(2.7-3.0)
Zweers	92	P,W,B	3846	S/O	Y		21								5 of 5 gr. of S	1.5-2.1
Brasche	99	P,W													3 of 7 S	(1.4-1.4)
Hawkins	91	P	255		N	Y	6							6	S score	
Jaakkola	95	P,W,B	1828	O		Y/N			18		9				2 of 14 S	(1.3-1.7)
Jaakkola	95	P,W,B	1295	O		Y/N			18			2			1 of 14 S	(1.8-1.8)
Jaakkola	95	P,W,B	1519	O		Y/N			18				3			

^ as reanalyzed by Mendell (1990) #P = personal factors, W = work factors, B = building factors
*In mechanically-ventilated buildings **Hum = Humidification ^^ gr = groups



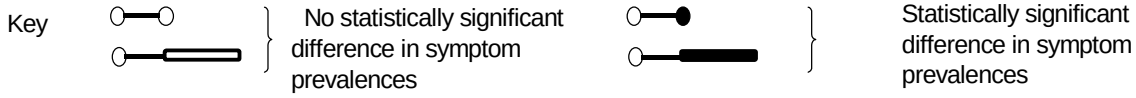
however, only ten of 540 rooms in this study had natural ventilation. In one of the five assessments (Skov et al., 1990) with increased symptoms in buildings with simple mechanical ventilation, two buildings with mechanical ventilation had humidifiers, a possible risk factor. When prevalences were significantly higher with simple mechanical ventilation, the odds ratios or relative risks ranged from 1.4 to 2.3, with one outlier of 6.0. One of these seven assessments had the opposite finding (significantly more symptoms with natural ventilation)

and one had no statistically-significant findings. In two other assessments in Table 2, prevalences of symptoms with mechanical exhaust ventilation did not differ significantly from prevalences with natural or simple mechanical ventilation.

Table 2. Comparisons of symptom prevalences among buildings without air conditioning.

Reference		Study and Building Characteristics					Ventilation System Type				Results	
First Author	Year	Controlled confounders#	No of respondents	Sealed (S) or openable (O) windows*	Smoking	Recirculation*	Natural Ventilation	Mechanical exhaust	Simple mechanical	Simple mechanical, with humidification	Symptoms with signicantly higher prevalences in assessment^^	Range of risk ratio or (odds ratio) for outcomes
Jaakkola	95	P,W,B	456	O		N	7	2				
Skov	90	P,W	2369	O		Y/N	9		5		2 of 2 gr. of S	1.4 - 1.8
Jaakkola	95	P,W,B	1460	O		Y/N	7		18		1 of 14 S	2.2
Mendell	96	P,W	300	O	N	Y	3		3		4 of 7 S	1.5 - 5.4
Burge^	87	none	1386	S/O			11		7		3 of 10 S	(0.7- 0.8)
Sundell	94	P,W,B	778	S/O								
Zweers	92	P,W,B	3009	S/O	Y		21				2 gr. of S	1.3 - 1.5
Sundell	94	P,W,B	788									
Zweers	92	P,W,B	2879	S/O	Y		21				4 gr. of S	1.4 - 2.1

^as reanalyzed by Mendell 90 #P = personal factos, W = work factors, B = building factors
*In mechanically-ventilated buildings **Hum = Humidification ^^gr. = group



The results portrayed in Tables 1 and 2 provide minimal information on the potential additional risks of humidification. Hedge et al. (1989) compared symptom prevalences among three sets of air-conditioned buildings: buildings without humidification, buildings with steam humidification, and buildings with evaporative humidification. The prevalences of five of ten symptoms differed significantly among the three HVAC types; suggesting that humidification type may affect symptom prevalences. For eight of ten symptoms, prevalences were highest with evaporative humidification. The results reported in Table 1 of Zweers et al. (1992), comparing symptom prevalences with simple mechanical ventilation (independently with and without humidification) to symptom prevalences with natural ventilation, also suggest that humidification may be associated with higher prevalences of two out of five symptom groups.

CONCLUSIONS

Relative to natural ventilation, air conditioning with or without humidification was consistently (16 of 17 assessments) associated with a statistically significant increase in the prevalence of one or more SBS symptoms. Prevalences were typically higher by roughly 30% to 200% in the air conditioned buildings. In two of three available assessments (from a single study), symptom prevalences were also significantly higher in air conditioned buildings than

in buildings with simple mechanical ventilation and no humidification. The available data also suggest, with less consistency, an increase in risk of symptoms with simple mechanical ventilation relative to natural ventilation. In five of seven assessments, SBS symptom prevalences were higher in buildings with simple mechanical ventilation with or without humidification than in buildings with natural ventilation or in sets of buildings with either natural or exhaust ventilation. Insufficient information was available for conclusions about the potential increased risk of SBS symptoms with humidification or with recirculation of return air. The statistically significant associations of mechanical ventilation and air conditioning with SBS symptoms are much more frequent than expected from chance. The consistent associations reported in this synthesis are not likely to be a consequence of confounding by personal or job factors, textiles, building age, indoor temperature, indoor humidity, depth of building bays, and dusty surfaces. All studies were performed in moderate or cold climates; thus, the findings reported in this paper may not apply for buildings in hot humid climates.

The reasons for the consistent increases in symptom prevalences with mechanical ventilation and particularly with air conditioning remain unclear. Multiple deficiencies in HVAC system design, construction, operation, or maintenance may contribute to the increases in symptom prevalences, including deficiencies that lead to pollutant emissions from HVAC systems.

ACKNOWLEDGEMENTS

This review was supported with a grant from the Academy of Finland and by the Assistant Secretary for Energy Efficiency and Renewable Energy, Office of Building Technology, State and Community Programs, Office of Research and Standards of the U.S. Department of Energy under contract No. AC03-76SF00098. The authors thank Dr. Mark J. Mendell and Mr. Hal Levin for their many insightful comments on a draft of this document.

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II. AIR FLOW AND POLLUTANT TRANSPORT MODELING

MODELING TRANSIENT CONTAMINANT TRANSPORT IN HVAC SYSTEMS AND BUILDINGS

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ABSTRACT
A mathematical model of the contaminant transport in HVAC systems and buildings is described. The model accounts for transients introduced by control elements such as fans and control dampers. The contaminant transport equations are coupled to momentum equations and mass continuity equations of the air. To avoid modeling variable transport delays directly, ducts are divided into a large number of small sections. Perfect mixing is assumed in each section. Contaminant transport equations are integrated with momentum equations in a way that guarantees mass continuity by using two non-negative velocities for computing the mass transport between elements. Computer simulations illustrate how the model may be used to analyze and design control systems that respond to a sudden release of a toxic contaminant near a building. By coupling transient flow prediction with transient contaminant prediction, the model overcomes a number of problems with existing contaminant transport codes.

INDEX TERMS
Air transport, HVAC, Modeling pollutant concentrations

INTRODUCTION

Contaminant transport indoors is influenced by the motion of indoor air (advection) and by diffusion. In buildings with mechanical ventilation, the movement of air by mechanical equipment plays a significant role in the transport of contaminants. Mechanical equipment such as fans and dampers continually adjust the flow rates of air in buildings, sometimes slowly, but other times abruptly.

Two kinds of computer codes exist for modeling contaminant transport indoors. One kind, called computational fluid dynamics (CFD), solves the partial differential equations governing mass, momentum, and energy transport on a fine grid. CFD codes are complex, expensive, and difficult to use, so simpler models have been proposed that use lumped or zonal models to predict contaminant transport. Examples of such zonal models are CONTAM (Walton, 1997) and COMIS (Fuestel, 1999).

Existing zonal models assume steady-state air motion. Airflows are determined by solving a set of nonlinear algebraic equations for air mass conservation at a set of zones. After the flows have been determined, mass transport of the contaminant is predicted by equations governing zone-to-zone mixing.

The model described in this paper is motivated by the need for a zonal model that can predict contaminant transport under conditions where the air motion is highly transient. The intended application is the prediction of controlled response following the release of a toxic contaminant in or near a building. If the release could be detected, then the building controls could be used to rapidly redistribute air to minimize injury. Existing zonal models are not suited to predicting rapid response.

The approach is to couple equations governing zone-to-zone mixing with equations of motion for the air. Transient behavior due to both mixing and unsteady air motion are solved simultaneously. Flow rates from the equations of motion are used in the advection terms of the mixing equations. Mass conservation of the contaminant is ensured by using two non-negative velocity terms in the mixing equations derived from the equations of motion.

METHODS

The equations of motion for airflow in ducts has been described elsewhere (Federspiel et al., 2001). A summary is provided here. Equations of motion are derived from momentum equations in HVAC system ducts. The change of momentum in a duct is equal to the sum of forces acting on the boundaries of a control volume draw at the surfaces and ends of the duct. Forces include pressure at duct end points, which are often junctions with other ducts, fluid friction, pressure drops across minor loss components such as elbows, and pressures developed by fans. The momentum equations for a network of ducts can be expressed by the following set of differential equations:

$$M\dot{V} = -F\langle V|V|\rangle - K\langle V|V|\rangle + A_f P_f + A_e P_e + A_j P_j \qquad (1)$$

where M is a mass matrix, V is a velocity vector, F is a matrix of friction terms, K is a matrix of loss coefficient terms, P_f is a vector of fan static pressures, P_e is a vector of external pressures, and P_j is a vector of junction pressures.

Conservation of mass at each junction in the network is described by the following set of linear equations:

$$A_j^T V = 0 \qquad (2)$$

Together, Equations 1 and 2 form a set of differential algebraic equations (DAE) with index 1. They can be reduced to a set of ordinary differential equations by differentiating the conservation constraint (Equation 2), substituting Equation 1, solving for the junction pressures, then substituting the junction pressures into Equation 1.

Contaminant transport in ducts and in rooms is modeled using mixing equations with advection from one zone to another. Figure 1 shows a schematic of a three-zone mixing system. To ensure conservation of mass of the contaminant two non-negative flow terms connect the zones.

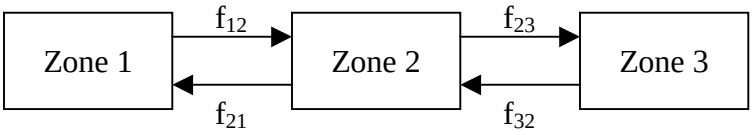

Figure 1: Schematic diagram of a three-zone mixing system.

The mixing equation for zone 2 is as follows:

$$\frac{d}{dt}(M_2\omega_2) = f_{12}\omega_1 - f_{21}\omega_2 - f_{23}\omega_2 + f_{32}\omega_3 \tag{3}$$

where M_2 is the mass of air in zone 2, ω denotes mass concentration, and f denotes mass flow rate. When a sequence of zones is used to model a duct, all of the flow terms in one direction equal the flow computed from the momentum equation, and all of the flow terms in the opposite direction are zero. Using the non-negative flow terms simplifies the switching in the mixing equations that results from flow reversals in the equations of motion, and adds very little overhead to the simulation code.

The transport delay resulting from contaminant transport in ducts is modeled by breaking the duct into a large number of adjacent sections in a so-called "tanks-in-series" model. As the number of sections approaches infinity, the behavior of the multi-zone mixing model approaches that of a plug-flow model. Figure 2 shows how well mixing models with increasing numbers of sections approximate a plug flow system.

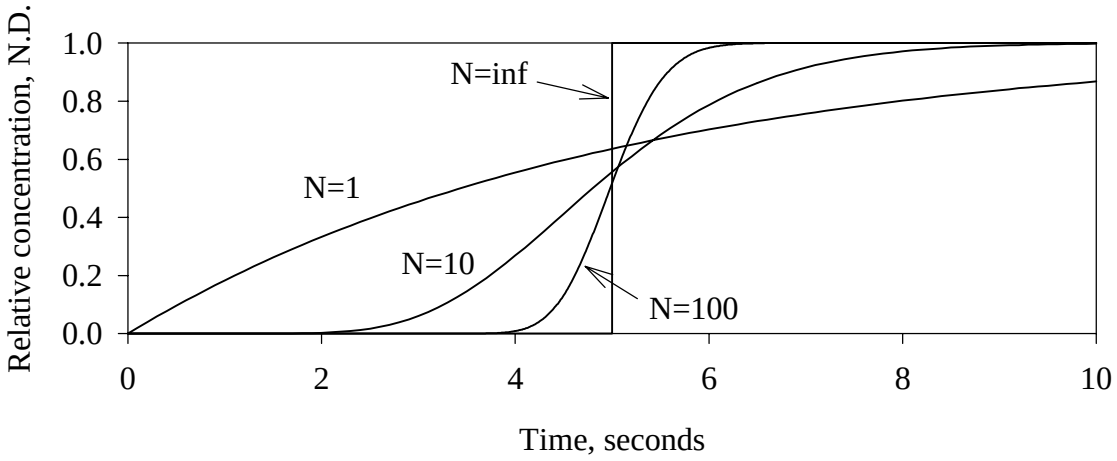

Figure 2: Approximation of plug flow by mixing systems.

RESULTS

In this section we show examples of simulations of a simplified system configured to model an air-handling system serving a single zone. The system has five ducts (outdoor, recirculation, supply, return, and exhaust), with the following diameters: 1.2, 0.7, 0.6, 0.5, and 0.4 meters, respectively. The duct lengths are 3, 5, 100, 80, and 3 duct diameters, respectively. The room is 11.5 by 23 meters in area, and is 3 meters high. The system has a supply and return fan, and control dampers in the outdoor, recirculation, and exhaust ducts.

We modeled a scenario where a contaminant suddenly enters the outdoor air intake at a fixed concentration of 345 μg/kg. We assumed that the contaminant would enter the exhaust duct at that concentration if the flow there reversed, and that the contaminant would enter the room by infiltration at that concentration if the room pressure became negative. When the contaminant first enters the system a control action is taken. We modeled two control actions. The first is the standard shutdown action for an air-handling unit. The fans shut down together, the outdoor and exhaust dampers close, and the return damper opens. We also considered an alternative action in which the return damper closes in order to reduce air motion as quickly as possible. The damper actuators move at a fixed speed of one degree per second. The fans slow down exponentially with a time constant of 90 seconds.

Figure 3 shows the room concentration as a function of time for both actions. The standard shutdown procedure is more effective at controlling the concentration of the contaminant. While closing the return damper clearly reduces the supply flow rate as shown in Figure 4, it also increases the outdoor airflow rate (Figure 5), increasing the concentration in the supply duct (Figure 6).

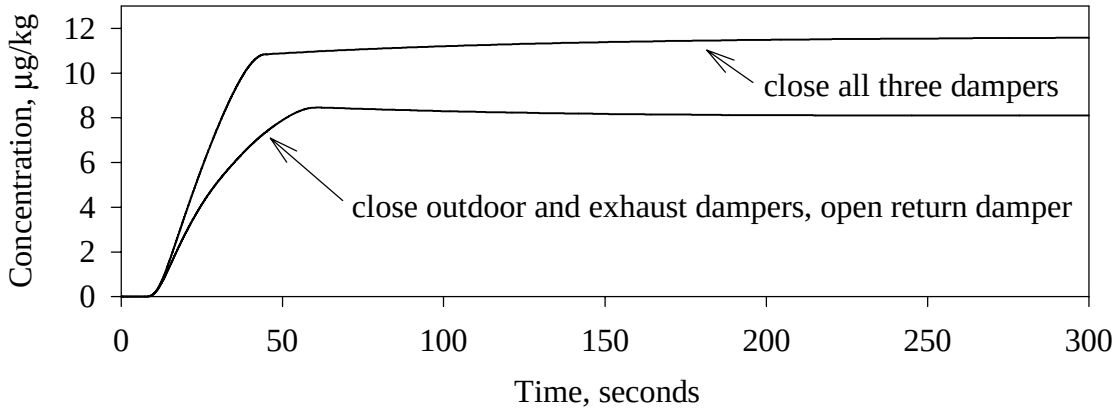

Figure 3: Room concentrations for both actions.

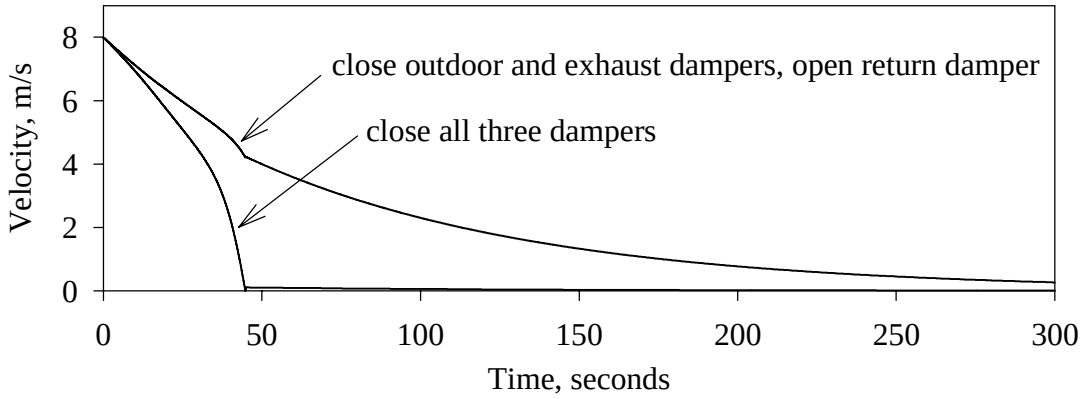

Figure 4: Velocity in supply duct for both actions.

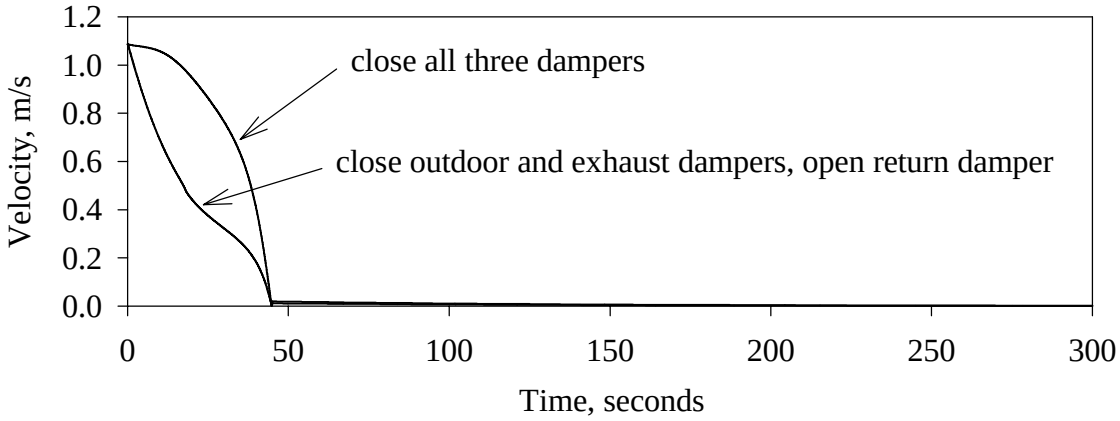

Figure 5: Velocities in outdoor air duct for both actions.

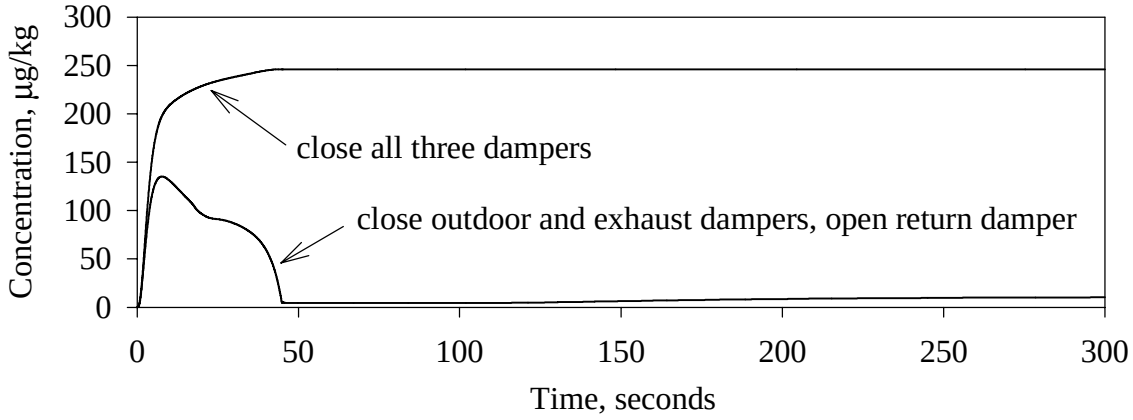

Figure 6: Concentration at the inlet of the supply duct for both actions.

DISCUSSION

The model proposed in this paper was developed specifically to analyze and design control systems that respond to accidental or intentional releases of toxic contaminants. The simulations in the previous section illustrate how it can be used for that purpose. However, it could also be used to study how the normal transient behavior of HVAC controls impacts indoor air quality. For example, the model could be used to assess how start-stop operations of small, packaged equipment impact the indoor concentration of normally occurring volatile organic compounds indoors.

The model described in this paper may require a solver that can handle stiff systems, which are systems that have a wide range of eigenvalues. The momentum equations will be stiff if some ducts are very short relative to others or if the loss coefficients in some ducts are very high. In practice both of these conditions are common. Furthermore, the contaminant transport equations will be stiff if zones of widely varying sizes are used. This is likely since some zones will represent duct sections while others will represent rooms. To get acceptable

behavior from the simulations in the previous section it was necessary to use a solver designed to handle stiff systems, to tighten the tolerances, and to force it to use no more than second-order integration, which is absolutely stable.

The model proposed in this paper could help overcome some of the significant problems associated with zonal models such as CONTAM and COMIS, which rely on fairly limited types of steady-state flow models. Using transient models provides one way to escape the modelling restrictions in these programs. Furthermore, for airflow applications it is trivial to provide consistent initial conditions for the simulation simply by setting all velocities and fan speeds to zero. Therefore it is always possible to arrive at a solution to the airflow system using transient models, even if the modeller is only interested in steady-state behavior.

Another problem associated with zonal models that rely on steady-state flow is that the models assume that contaminants move instantaneously between rooms. They don't normally treat duct sections as zones, so they don't account for the fact that the HVAC system itself can store contaminants. This means that they may over-predict concentrations after events such as the kind show in the simulations in this paper.

The momentum of fan wheels and air mass in ducts causes air to continue to move even after fans have been de-energized or fan speeds changed. If existing zonal models are used to predict transient behavior, then errors may be made in predicting flows because fan speed lags fan commands and air velocity lags fan speed. The model in this paper overcomes this problem.

CONCLUSION AND IMPLICATIONS

We have developed a model that predicts the concentrations of contaminants in HVAC systems and buildings during sudden transient conditions. It differs from existing zonal models by simultaneously predicting unsteady airflow and unsteady contaminant transport. The model can be used to analyze and design control actions taken in response to an accidental or intentional release of a toxic contaminant in or near a building.

ACKNOWLEDGEMENTS

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MIXING OF A POINT-SOURCE INDOOR POLLUTANT: NUMERICAL PREDICTIONS AND COMPARISON WITH EXPERIMENTS

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ABSTRACT
In most practical estimates of indoor pollutant exposures, it is common to assume that the pollutant is uniformly and instantaneously mixed in the indoor space. It is also commonly known that this assumption is simplistic, particularly for point sources, and for short-term or localized indoor exposures.

We report computational fluid dynamics (CFD) predictions of mixing time of a point-pulse release of a pollutant in an unventilated mechanically mixed isothermal room. We aimed to determine the adequacy of the standard RANS two-equation *(k-ε)* turbulence model to predict the mixing times under these conditions. The predictions were made for the twelve mixing time experiments performed by Drescher et al. (1995). We paid attention to adequate grid resolution, suppression of numerical diffusion, and careful simulation of the mechanical blowers used in the experiments. We found that the predictions are in good agreement with experimental measurements.

INDEX TERMS
Air transport, Microclimates, Exposure assessment, Air and pollutant transport modeling and measurement, Modeling indoor pollutant concentrations.

INTRODUCTION
Indoor air quality investigations usually assume a uniform distribution of pollutants throughout the interior space. This assumption of instantaneous mixing offers some compelling advantages in domain of its validity. For experimental purposes, the measurement of the concentration at only one point can be used to obtain the overall concentration throughout the room. In modeling studies, the assumption leads to governing equations that are either systems of ordinary differential equations or algebraic equations, whose numerical solutions are straightforward in contrast to the partial differential equations that must be solved if one takes real mixing into account.

However, particularly for point sources, and for short-term or localized indoor exposures, this assumption proves too simplistic for the initial period of the mixing of the pollutant in the room air. Lambert *et al*. (1993) showed, for example, that the levels of respirable suspended particles and nicotine were respectively 40% and 65% lower in no-smoking sections of restaurants than in the smoking sections. The mixing problem has two aspects: (1) how to determine the conditions under which the well-mixed approximation is inappropriate, and (2) how to model pollutant concentrations when the well-mixed approximation is inappropriate. We earlier addressed the first question experimentally for rooms mixed with natural or forced convection (Baughman *et al*. 1994, Drescher *et al*. 1995). A “mixing time” is defined for a

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point pulse release of a pollutant in the room, under particular flow conditions. At times beyond the mixing time one can safely use the well-mixed assumption. At times earlier than the mixing time, the well-mixed assumption is a poor approximation.

Previous work on modeling imperfectly mixed pollutant concentrations aimed for the introduction of an additional mixing factor (Hoegg, 1972) (Ishizu, 1980). More recently, a two-compartment model was applied, artificially defining a small virtual space around the source (e.g., an area of workplace pollutant emissions) also with a uniform concentration, but different from that in the rest of the room (Furtaw, 1996). Although construction allowed for initial concentration build-up near the source, comparisons with experiments showed that concentrations near the source exceeded the predictions of this modeling approach. In the current work, we focus not on improving approaches to model exposure under poorly mixed conditions, but on determining the mixing time beyond which the well-mixed assumption is valid, and before which it is a poor approximation.

Computational fluid dynamics (CFD) solves the partial differential equations governing pollutant mixing and can provide solutions with high spatial and temporal resolution. With the improvements in CFD methods and increased computer power, CFD is increasingly common in research. Nevertheless, making accurate CFD predictions of room airflow remains a difficult territory, requiring great care in defining the settings and numerical properties of the model.

Most of the previous work on the CFD simulation of contaminant dispersion in a room was performed without corresponding experiments (Baker and Kelso, 1990). Gadgil *et al*. (2000) provide a brief review. Yaghoubi *et al*. investigated mixing of pollutants using simulations with varied emissions from a pollutant source, cooling and heating locations, and incoming air temperatures (Yaghoubi *et al*., 1995). Roy *et al*. (1994) used a CFD code to predict contaminant dispersion in a kitchen-hood geometry.

The specific goal of this research was to explore the adequacy of the standard (k-ε) turbulence model for predicting the mixing time of a point-source release, by comparing CFD predictions with experiments releasing a point pulse of Carbon Monoxide (CO) in an isothermal room with forced convection airflow. We simulated a series of 12 experiments releasing a pulse of CO from a point-location into a mechanically mixed unoccupied room, carried out by Drescher during her Ph.D. thesis (Drescher, 1994).

MIXING TIME THEORY

The characteristic mixing time t_{mix} is defined as the interval from the time of point-pulse release of the pollutant into the room to the time at which the standard deviation of the pollutant concentration $C_i(t)$ (measured at N monitoring points in the room, i=1,…,N) drops permanently below 10% of the arithmetic mean concentration $\overline{C}\,(t)$ of all sensors.

In the Drescher series of experiments, N=9 sensors were used, selected such that the mixing time calculated from their monitored values is the same as that calculated from a denser network of 41 sensors used by Baughman *et al*. (1994). Since the CO injection was not truly instantaneous (it took 20 seconds), the time interval was measured from the midpoint of the duration of CO injection. Engineering analysis shows that under conditions of forced convection, t_{mix} should be proportional to the inverse of the cube root of mechanical power P, deposited into the room air by the array of blowers.

$$t_{mix} = c * \frac{M^{1/3} L^{2/3}}{P^{1/3}} \qquad (1)$$

where c is the constant of proportionality, M denotes the mass of the air in the room and L is a characteristic dimension of the room. The term on the right-hand side is also known as power parameter p, and has units of time.

GEOMETRICAL SETTINGS

Figure 1 displays the schematic for the experimental room, which measured 3.53m x 3.74m x 2.36m high (31m^3). Metal foil covered the windows to block sunlight. The temperatures at the walls and at different heights in the middle of the room differed from the average temperature by amounts ranging from 0.25 to 1 Kelvin with an average of 0.59 Kelvin. The influence of natural convection was neglected in the CFD simulations, and we apply a correction to experimental mixing time to remove the small influence of natural convection, before comparison with predictions. Five independently powered identical centrifugal fans, each fitted with a 60 cm long plastic exhaust pipe, were placed 3 cm above the ground with their plastic exhaust pipes parallel to one another and to the floor. At the start of each experiment, 1.5 l of pure CO was released over a period of 20 seconds through a perforated syringe. The CO concentrations at various locations in the room were monitored with nine sensors, and each of the twelve experiments was repeated once.

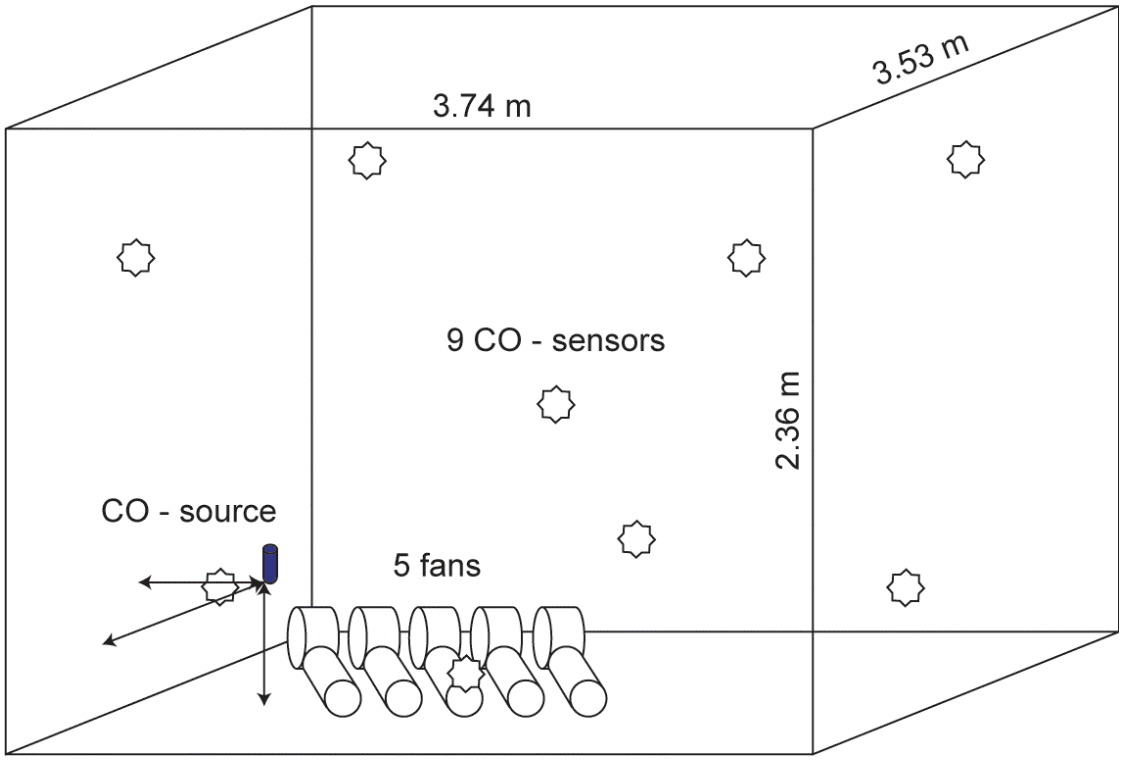

Figure 1. Schematic model of the experimental room

In the CFD simulations, the physical space occupied by the blowers (which refers to the fans and pipes together) was excluded from the computational domain. Appropriate boundary conditions modeled the blower intakes and exhausts. Pollutant mass leaving the computational space by entering a blower intake immediately reappeared at the blower exhaust and re-entered the computational space. Infiltration through the walls, experimentally

measured to range from 0.03 to 0.08 air changes per hour, was ignored. A small area on the room surface was defined as a pressure boundary to improve stability of the solution procedure. This allowed a small (but negligible) amount of pollutant to leave the room during the simulated experiments. The syringe was simulated as the combined surface of the holes ($12cm^2$), releasing CO at a speed of 6.29 cm/s.

NUMERICAL METHODS

Most of the room space was discretized with a coarse 10x10x10 cm mesh, except near the boundaries and near the blower jets. An inner large block of cells surrounding and enclosing the blowers was aligned with the exhaust pipes of the blowers to avoid imposing the grid orientation on the exhaust jets. The final working grid had 130,942 nodes. The fans and attached pipes were simulated using block structures with 5mm resolution at the ends of the blower pipes where the highest velocities were reported. The walls of the room and the outside of the blowers were defined as impermeable non-slip boundaries (except for the small pressure boundary area mentioned above). The blower inlets and exhausts were defined to have prescribed flows with fixed velocity profiles and with pollutant concentrations coupled to each other through user-subroutines. The grid quality was verified by testing the resulting predictions for independence of the grid size, time-step and turbulence parameters. A constant time step size of 0.1 sec was used for each simulation.

The Navier-Stokes equations were solved using a commercial code based on a finite-volume fully implicit method and selecting the standard (high Reynolds number) RANS two-equation *(k-ε)* turbulence model. In this work, the MARS (Monotone Advection and Reconstruction Scheme) differencing scheme was chosen, because it is second-order accurate. The solution algorithm applied was SIMPLE (Semi Implicit Method For Pressure Linked Equations) for the calculations of the steady-state velocity fields (without pollutant) and PISO (Pressure Implicit Split Operator) for the transient calculations of the pollutant dispersion in the room (with fixed velocities and turbulence parameters).

The chosen convergence criterion was that the residuals for all calculated dependent variables decreased at each cell below a value of 10^{-4} for the steady state calculations. Iterations to reach convergence for the steady state ranged from 1817 to 4855, requiring 10 to 35 hours processing time on a server with 2GB RAM and two processors. The transient calculations required half as much processing time.

COMPUTATIONAL RESULTS

Drescher *et al*. performed 2 replicates of 12 experimental set-ups. The number of blowers operated and the average velocity of the air jets at the blower exhausts (and therefore also at the inlets) varied from one setup to another. CFD computations were performed for all 12 setups. Three additional computations were performed to obtain predictions for a gap left in the experimental dataset regarding the power parameter values. Since the simulations were performed isothermally, the experimental data were corrected for the small influence of natural convection before comparison with simulation results. The total mechanical power deposited into the room air by the blower jets ranged from 0.001 W to 0.985 W.

Typical predictions of concentration over time for the nine different sensors displayed a high initial peak concentration, as the plume of pollutant passed each sensor for the first time, and several smaller secondary peaks from recirculation of the plume, until the concentration at all sensors locations converged toward the fully mixed state. Further details are omitted for brevity.

COMPARISON WITH EXPERIMENTS

Detailed experimental data were available about the fully developed turbulent velocity fields at the blower pipe exhausts, which was essential in simulating them correctly. Data from continuous temperature measurements at each wall supported our decision to conduct the simulations isothermally. The mixing times obtained from the experiments (two experiments per setup, adjusted for natural convection) and predicted from the simulations were compared as a function of the power parameter *p* (1), displayed in Figure 2.

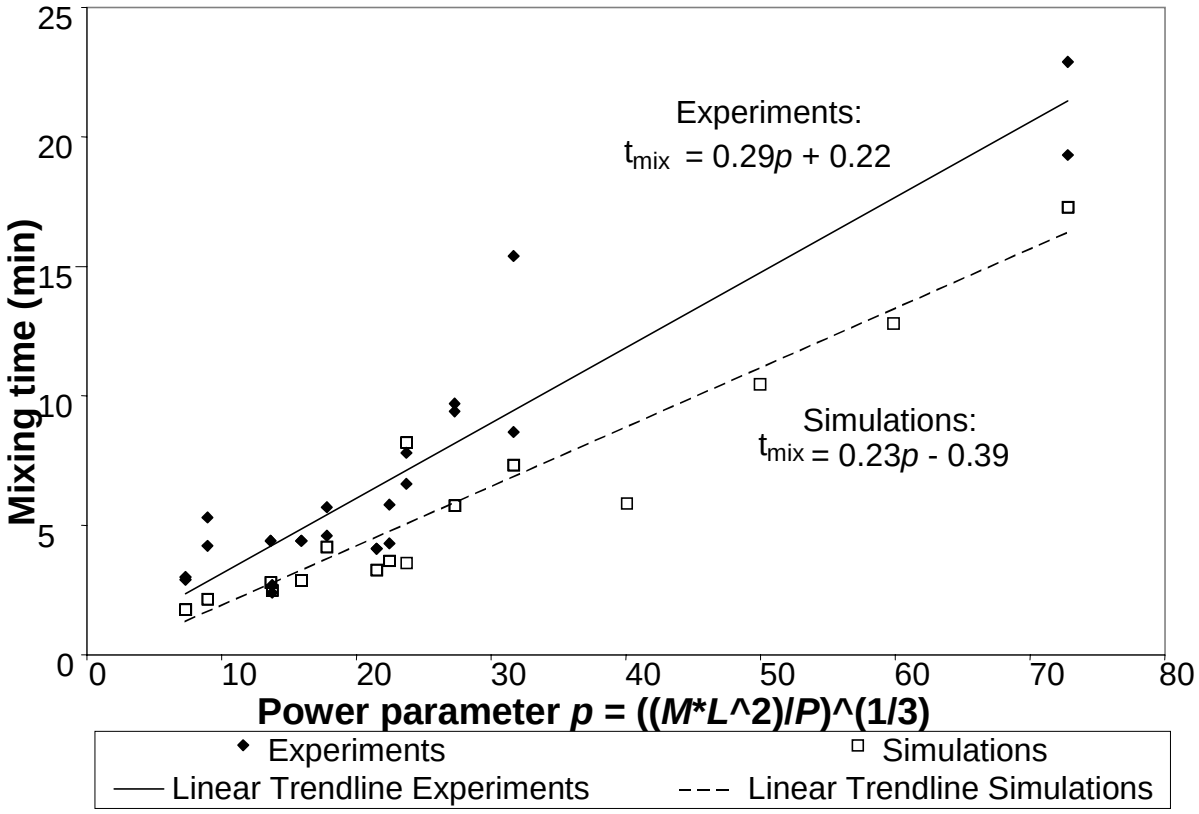

Figure 2. Mixing times as a function of the power parameter *p*, experiments and simulations. Each experiment was replicated twice, yielding two experimental points for each power. Experimental values are shown corrected for the small effect of natural convection.

DISCUSSION

As can be seen in Figure 2, there is significant variability in the experimental results. The two values of mixing time for identical setups, obtained from replicate experiments, differ on average by 18%. The constant of proportionality, *c*, between the power parameter and the mixing time (expressed in seconds, not in minutes as depicted in Figure 2) agrees reasonably well for experimental data and simulation results. For the experiments *c* is 15 ± 5.2, and for the simulations12 ± 3.0. These results demonstrate the ability of the High-Reynolds number *(k-ε)* turbulence scheme to adequately predict the experimental mixing times, under the conditions tested in the setups.

We also find a good correspondence between the mixing times of the simulations and the turbulent parameters averaged over the flow field. Again, details are omitted for brevity.

CONCLUSION AND IMPLICATIONS

In this work, we found that the standard *(k-ε)* turbulence model, using a carefully generated grid, carefully designed boundary conditions and the MARS solution algorithm, suppressing numerical diffusion, gives mixing time predictions within 15% of experimental measurements over an order of magnitude range in mixing time.

In future work, it would be useful to explore the capability of a low-Reynolds number *(k-ε)* turbulence model to predict mixing times, and also to determine the extent to which the mixing time value is a property of the given airflow setup in the room, and whether it is independent of the location of the point pulse release. Finally, predictions of mixing time under non-isothermal conditions also need to be explored and tested against experimental data.

ACKNOWLEDGEMENTS

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ASSESSING MULTIZONE AIRFLOW SIMULATION SOFTWARE

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ABSTRACT
Several standard multizone modeling programs, in order to improve their computational efficiency, make a number of simplifying assumptions. This paper examines how those assumptions reduce the solution times and memory use of the programs, but at the cost of restricting the models they can express. Applications where these restrictions may adversely affect the program's usefulness include: (1) natural ventilation, when buoyancy effects dominate mechanically-driven flow; (2) duct system design, when losses in T-junctions affect the system performance; and (3) control system design, when the dynamic transport of pollutants plays a significant role in the simulated system.

INDEX TERMS
COMIS, CONTAM, Modeling, Multizone, Pollutant transport.

INTRODUCTION
Multizone models form the basis of most computer simulations of airflow and pollutant transport in buildings. Whole-building analysis relies on these lumped-parameter models, rather than on fluid-dynamic models, due to the high demand the latter techniques make on computer memory, processing power, and input data.

The design of the current generation of multizone programs results from a systematic drive to improve their computational efficiency, mainly by the introduction of specialized algorithms and data structures. This specialization has allowed engineers and scientists to explore a wide range of building airflow problems. However, with increased understanding of these problems has come the desire to explore questions that lie beyond the capabilities of current multizone tools.

This paper considers two popular and well-characterized multizone simulation programs, CONTAM (Walton, 1997) and COMIS (Feustel, 1999). The paper examines how design decisions made to improve their efficiency also restrict their ability to incorporate a number of interesting, and potentially important, airflow and pollutant transport models. After describing the computational approach taken by the programs, this paper describes some new models that would be difficult to implement under the current software architecture. These models would prove useful in a number of application areas, including the study of natural ventilation and the design of novel mechanical ventilation systems.

The decision to focus on CONTAM and COMIS follows from their popularity, and the author's familiarity with them, rather than an assessment that they are either more or less capable than other multizone programs. Furthermore, the comments made here pertain to the extremes of these programs' capabilities, and do not imply that the codes perform poorly when applied to their intended use.

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COMPUTATIONAL APPROACH

Multizone models idealize a building as a collection of well-mixed spaces, or *zones*, connected by discrete flow paths. In CONTAM and COMIS, the user constructs a building description by assembling various component models, each representing a zone or a flow path. A simulation predicts the system's behavior based on the interactions of the assembled components. Flow path models range from simple cracks, to windows and doors, to active elements such as fans. By contrast, the programs provide only one zone model, which represents a well-mixed space with simple hydrostatic pressure variations.

Both CONTAM and COMIS decouple the airflow problem from the thermal problem (finding the temperatures in the zones) by assuming the temperatures remain fixed, for example at user-supplied values. This yields a steady-state airflow system, which can be embedded in a dynamic thermal system by treating the resulting airflows as constant until the next simulated time, when new zone temperatures produce a new steady-state airflow solution.

Similarly, the programs decouple the airflow and pollutant transport problems, by assuming the amount of pollutant in a space does not affect the airflows. Hence they idealize a building as composed of: (1) a steady-state airflow network, defined by algebraic equations which, given the zone temperatures, describe the mass conservation of air; and (2) a dynamic pollutant transport model, defined by ordinary differential equations which, given the airflows, describe the mass conservation of pollutants.

This paper mainly concerns design features in the software that solves the airflow equations. Both programs represent the state of the airflow system using the pressures in the zones. Each zone has a single reference pressure, chosen by the airflow solver. The computation proceeds by: (1) finding the pressures at the points where the flow elements connect to each zone, based on the zone reference pressures and temperatures; (2) finding the flows in each path, based on the pressures and densities of air at its terminals; and (3) summing the flows in and out of each zone. The solver adjusts the reference pressures to achieve mass balance, in other words, so that the mass flow rate of air into each zone matches the flow rate out of the zone. The program then passes these flows to the pollutant transport solver.

MODELING RESTRICTIONS

This section describes some design decisions taken in CONTAM and COMIS in order to improve the efficiency of solving the steady-state airflow network. A more complete analysis, given by Lorenzetti (2002), details the computational advantages gained by each decision. These include improved calculation speed, reduced computer memory requirements, and convergence guarantees. It is important to bear in mind that many of these decisions were made to bring multizone simulation to desktop computers with less capacity than those of today.

Probably the most important decision was to restrict the component models in a way that produces symmetric Jacobian matrices (Lorenzetti, 2002). The Jacobian stores the derivatives of the nonlinear equations that define the airflow network. At each time step, CONTAM and COMIS repeatedly calculate and factor new Jacobian matrices, extracting trial solutions for the airflow system using variations on Newton-Raphson's method. A symmetric matrix requires only about half the storage, and may be factored in about half the time, as a general full matrix (Dennis and Schnabel, 1996). In fact, the specialized sparse matrix techniques in

CONTAM and COMIS run even faster than this (Walton, 1997). These efficiency gains are important since matrix factorization dominates the solution time for the nonlinear system.

In practical terms, a symmetric Jacobian requires that each flow element: (1) connect to exactly two zones; and (2) calculate the airflow as a function of the pressure difference between those zones. These requirements follow from the fact that the Jacobian expresses the derivatives of the mass flows with respect to the zone reference pressures. Symmetry implies that the flow through a path must not change if the reference pressures in the zones it connects both change by the same amount.

Another important design decision was to require positive-definite system matrices. This requirement was adopted because a system that always produces symmetric positive-definite Jacobians must have a unique solution (Lorenzetti, 2002). Furthermore, relatively simple search algorithms can always find that solution. Finally, a positive-definite matrix simplifies the factorization algorithm still further.

In practical terms, a positive-definite Jacobian requires that: (1) each flow element gives more positive flow with more positive pressure drops in the flow direction; and (2) each zone connects, directly or indirectly, to at least one zone of known pressure. Most real flow elements satisfy the first criterion (Lorenzetti, 2002). The second requirement relates to the building topology, rather than to the component models individually.

The restrictions described above ensure the airflow systems have desirable mathematical properties. In addition, CONTAM and COMIS simplify the flow element models for convenience of implementation. Specifically, when they calculate the mechanical energy "lost" in each flow path, they do so independently of the flow path calculations. This "lost" energy represents the conversion of mechanical to thermal energy by viscous dissipation. It is a primary component of the pressure drop that forces air through the flow path. However, that pressure drop also depends on changes of potential energy (due to changes of height) and kinetic energy (due to changes of the velocity profile of air) through the flow path (McQuiston and Parker, 1988).

By calculating this lost energy outside of the flow path model, the programs simplify the code. In particular, imposing a global assumption about the energy balance frees the flow models from having to solve the energy and pressure-flow relations simultaneously. Unfortunately, this simplification prevents the model developer from representing more complicated relationships between the zone pressures, the density and velocity profiles of air in the path, and the mass flow of air through the path.

UNSUPPORTED MODELS

The design decisions made in CONTAM and COMIS make it difficult to implement some models, and impossible to implement others. This section describes some potentially useful and important models that the programs do not currently support. It identifies which models do not meet the fundamental restrictions imposed by the programs, and which could be implemented within the current framework, given sufficient programming effort.

The fact that each zone must have a single state variable (its reference pressure) precludes any momentum-balancing zone model. Thus no formulation based on the Navier-Stokes equations, such as the SIMPLE algorithm (Patankar, 1980), may be used to predict flow details within rooms. Note that it is possible to predict intra-room flows using the subzonal

models described by Wurtz et al. (1999), because these rely on temperature correlations and standard multizone flow elements, and do not introduce any new state variables to the airflow system.

Similarly, flow elements cannot incorporate momentum, since they must calculate steady-state flows based on the pressure difference between the zones they connect. Modeling momentum could be important in cases where a control system tries to quickly reduce the flow in a duct system, for example to shut off flow after detecting a pollutant in the supply air. In a real system, flow through a closing damper could be much greater than a steady-state model would suggest. Fortunately, in most applications a quasi-steady simulation, which approximates the dynamics by running a fan or damper model through a series of steady states, probably suffices.

Another dynamic of interest in duct systems, and one of potentially greater significance, concerns the delays associated with pollutant transport in flow paths. CONTAM and COMIS do not directly model transport delays, which would require them to account for both the pollutant mass stored in the flow paths, and the time needed to carry it between zones. Modeling all transport as instantaneous simplifies the assembly of the defining equations, but over-predicts the speed at which pollutants spread through a building. Fortunately the user may force the program to account for transport delays, by breaking up a flow path into a number of subpaths, connected in series between zones of appropriate volume. However this technique is less than ideal, as it complicates the model's input requirements, and unnecessarily increases the number of equations in the airflow system.

Restricting flow elements to depend on the pressure drop between two zones precludes proper modeling of duct T-junctions. Junctions, as three-port flow elements, cannot be defined solely in terms of pressure differences, because the mechanical energy dissipated in each branch of a junction depends on the flows in the other branches (McQuiston and Parker, 1988). Hence, they destroy symmetry in the system Jacobian. Modeling T-junctions properly would be very costly, in both storage and factorization times for the system matrices. Unfortunately, no simulation that demands fidelity in the duct system representation, for example as part of a duct design procedure, can ignore the pressure-flow characteristics of junctions. First, fitting losses usually exceed losses in a straight pipe. Second, junctions have unique characteristics among fittings. For example, changing the flow in one branch can even produce suction on another branch that previously faced a retarding pressure.

The fact that CONTAM and COMIS decouple the energy balance from the flow element relations means they cannot properly model buoyancy-driven bidirectional flow across a horizontal partition. For small pressure differences across the gap, the flow model must solve for the dissipated energy and the flow simultaneously, but the program design prevents this. This becomes important in, for example, a stairwell or elevator shaft, where temperature differences between floors of a building can set up recirculation between floors (Edwards and Irwin, 1990). In cases of natural ventilation, or forced ventilation where the mechanical system design tends to isolate floors, the programs may not predict two-way flows between floors. Note however that they can accurately model unidirectional, mechanically-driven flows in stairways (Feustel et al., 1985).

CONCLUSION AND IMPLICATIONS

Design decisions made in CONTAM and COMIS prevent them from implementing a number of potentially useful models. These include: (1) certain types of dynamic behavior; (2)

detailed duct systems; (3) pollutants that do not satisfy the well-mixed assumption; and (4) mixed natural and forced convection across horizontal partitions.

Some of these limitations derive from fundamental structural issues, like the reliance on symmetry-preserving flow elements. Others result from implementation details that could be resolved within the existing framework of the programs, such as the decoupled energy balance relation.

For users who wish to apply these programs to certain types of problems, these limitations may increase the need to modify or replace the programs. In particular, those interested in novel ventilation control systems, duct system design, or natural or hybrid ventilation, will find the programs' limitations outweigh their strengths as environments for simulating whole-building airflow and pollutant transport.

An effort should be made to identify and test alternate simulation environments. Desirable features include: (1) the ability to mix algebraic and differential equations when defining component models; (2) event-based models, for example to simulate a control system whose mode changes based on data from a pollutant sensor; (3) general nonlinear equation solvers, including a variety of direct and indirect matrix techniques; (4) an intuitive user interface for connecting component models in order to build up systems; and (5) support for wholly modular components, that is, an architecture that keeps the code for component models separate from the code that performs the simulation.

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MODELING THE SPREAD OF ANTHRAX IN BUILDINGS

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ABSTRACT
The recent contamination of several U.S. buildings by letters containing anthrax demonstrates the need to understand better the transport and fate of anthrax spores within buildings. We modeled the spread of anthrax for a hypothetical office suite and estimated the distribution of mass and resulting occupant exposures. Based on our modeling assumptions, more than 90% of the anthrax released remains in the building during the first 48 hours, with the largest fraction of the mass accumulating on floor surfaces where it is subject to tracking and resuspension. Although tracking and resuspension account for only a small amount of mass transfer, the model results suggests they can have an important effect on subsequent exposures. Additional research is necessary to understand and quantify these processes.

INDEX TERMS
Resuspension, tracking, deposition, multizone air flow, bio-aerosols

INTRODUCTION
The release and subsequent dispersion of anthrax spores within several U.S. buildings have illustrated the lack of detailed knowledge about the mechanisms by which building occupants can be exposed to anthrax or other bio-aerosols. In particular, the degree to which contamination spreads from one room to another – and by what mechanisms – is poorly understood quantitatively. The transport and fate of these bio-aerosols has implications not only for exposure assessment but for subsequent decontamination efforts as well.

The behavior of aerosols indoors has been studied both experimentally and theoretically (see, for example, Lai and Nazaroff, 2000; Thatcher et al., 2002; Wallace 1996) and many ofthe factors influencing aerosol transport and fate are reasonably well understood. These include: (1) air flow from room to room or between the building interior and outdoors driven by HVAC operation, by thermal stack effects, and by wind loads on the building shell; (2) deposition to interior surfaces; (3) filtration; and (4) coagulation. Occupant-related processes can also affect aerosol behavior. Some studies have observed differences between 'personal' and 'room' exposures, attributed – at least in part – to resuspension of deposited or tracked materials (Ozkaynak et al., 1996). However, these processes are not well quantified.

Our overall goal in this study is to examine the exposure potential from an anthrax release in a room using a broadly applicable mechanistic model. The objectives of this work are to gain a better understanding of the factors that influence potential exposures and to identify key features of this problem for which additional research is necessary.

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METHODS

Model Description

The basic elements of the aerosol transport and fate model are illustrated in Figure 1. The simulation program represents the contaminant transport network as an assemblage of storage locations, which accumulate contaminant mass, and transport elements, which carry contaminant between locations. The resulting system of coupled ordinary differential equations describes mass conservation in the storage locations. This approach allows the addition of other storage locations or transport processes not captured in this initial effort.

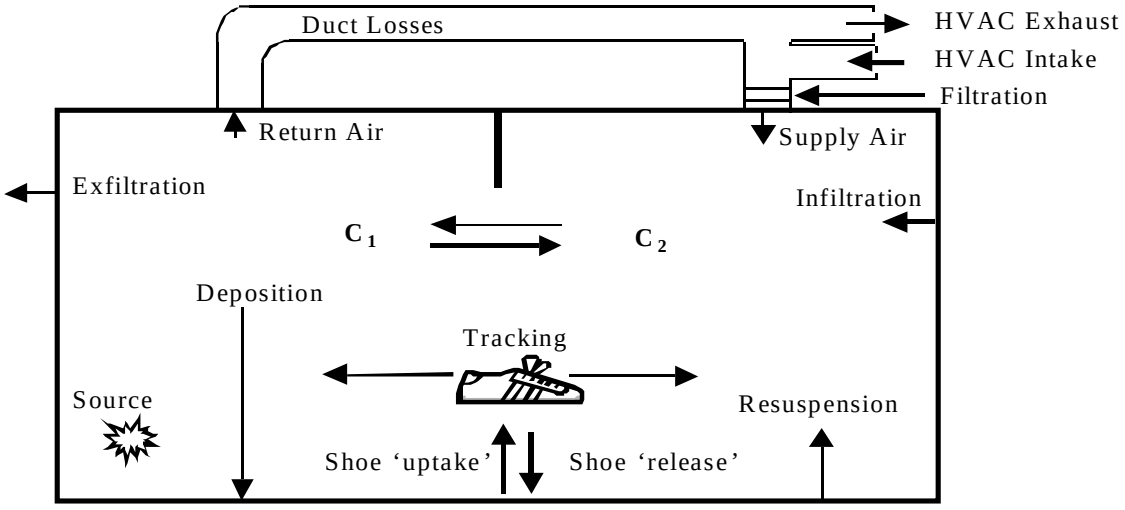

Figure 1. Schematic of the various anthrax transport and fate mechanisms. The floor surfaces are subdivided into tracked and untracked areas.

Storage locations include: (1) zones, which contain airborne contaminant (e.g. C_1 and C_2 in Figure 1); (2) zone and duct surfaces, such as walls and floors, where contaminant may settle; and (3) filters, which trap contaminants in the HVAC flow path. Transport elements include: (1) flow paths, which move air between zones; (2) deposition to zone and flow path surfaces; (3) a source rate term; and (4) an activity model, described below. For this study, the steady airflow rates were calculated using the multizone airflow program COMIS (Feustel 1999). .

The activity model accounts for three effects associated with human activity: (1) exchanges of contaminant between a person and building surfaces, for example between shoes and floor surfaces; (2) resuspension of contaminant from building surfaces to zone air, as a result of a person touching the surface (in this case, a person walking); and (3) tracking from room to room, for example when shoes load with pollutant from a heavily-contaminated floor, then lose mass after moving to a different, relatively clean floor. The exchange and resuspension processes vary directly with the activity level, defined as the rate at which a person touches building surfaces. The activity rate, the building surface for exchange, and the zone for resuspension, all may be scheduled within the model to vary over time. In this study, we partitioned floor surfaces into tracked and untracked areas. Deposition of airborne particles occurs uniformly on surfaces in both categories, however, tracking and resuspension occur only on the tracked surfaces.

Scenarios

We established a set of scenarios, built around a specific arrangement of rooms in a hypothetical office building, to explore the model assumptions and the various input

parameters. Figure 2 shows the floor plan of the office suite and the location of the anthrax source. The scenarios consist of different HVAC system configurations, building operating conditions and sets of occupant activity patterns. For the present paper we describe the results obtained from one scenario. In this case, the HVAC supply and return flows in each room are balanced, with a total HVAC system flow equal to 6 volume changes per hour and with the outdoor air supply making up 25 percent of this flow.

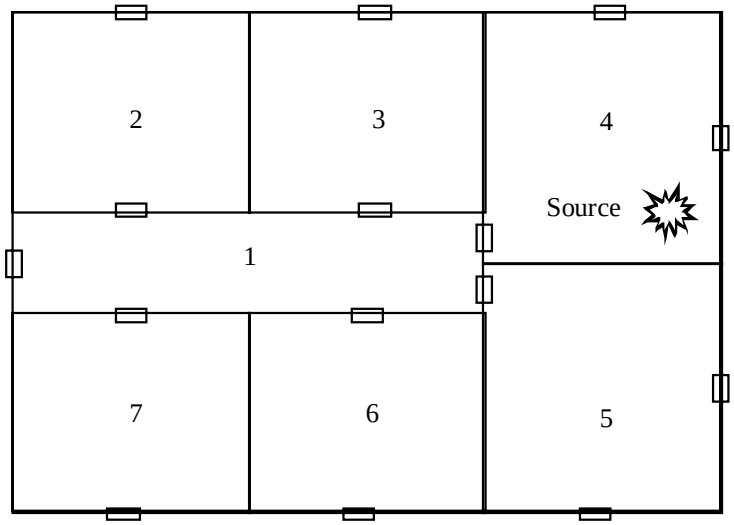

Figure 2. Floor plan of the hypothetical office suite. Room 1 is a hallway, room 2 is a common room, and rooms 3 to 7 are each assigned an occupant. The small open rectangles represent closed exterior windows and open doors between the offices and the hallway. The volume of each room is ~80 m^3.

We established activity patterns for six people in the space – each following a different movement pattern. For this scenario we assigned one person to each of the five offices and assumed they are all in their own offices at the time of the anthrax release in Room 4 (each person is labeled with their respective office number in Figure 2). A sixth individual (referred to as person 8) walks into the office suite two hours after the initial release, when airborne concentrations have been substantially reduced. Thus the exposure experienced by this person is dominated by resuspension. We simulated a 48 hour time period following the anthrax release, assuming that all six people followed the same activity pattern on day 2 as on day 1, but without a second anthrax release.

For our simulations we postulated that the source is an envelope containing 1 gram of anthrax opened in Room 4. We assumed that 50% of the initial mass stays in the envelope (and thus warrants no further consideration here), 75% of the mass of anthrax released from the envelope deposits uniformly on the floor surface in Room 4 and the remaining 25% disperses immediately and uniformly into the room air. This represents the initial state for the modeling. Note that exposures described below will scale with alternative mass distributions.

Other key assumptions are: (1) anthrax spores deposited on 'untracked' surfaces, in the ducts or on the HVAC filter remain in those locations; (2) spores deposited on the floor do not change physically, that is, spores deposited with a given aerodynamic diameter maintain that diameter for purposes of resuspension; and (3) the rates for anthrax 'uptake' and 'release' by shoes are strictly estimates and have been set equal. The key aerosol parameters are shown in Table 1; for the baseline case, we used the parameter values for five-micrometer-diameter

aerosols. Anthrax aerosols, consisting of spore aggregates, range in diameter from ~2 to ~10 micrometers (Thatcher et al., 2000).

Table 1. Values for aerosol parameters used in simulations

	Aerosol diameter (micrometers)				
Transport values (units)	1	3	5	10	Reference
Shoe uptake (h^{-1})	0.1	0.1	0.1 (a)	0.1	Current estimate
Shoe release (h^{-1})	0.1	0.1	0.1 (a)	0.1	Current estimate
Resuspension (h^{-1})	$1.2x10^{-4}$	$1.9x10^{-3}$	$.8x10^{-3}$	$.4x10^{-2}$	Thatcher & Layton 1995
Deposition-floors (h^{-1})	0.1	0.6	2.0	8.1	Composite experimental
Dep'n - walls/ceiling (h^{-1})	0.1	0.4	0.8	0.9	Composite experimental
Duct loss fraction (m^{-1})	$4x10^{-5}$	$6.0x10^{-4}$	$1.9x10^{-3}$	$6.1x10^{-3}$	Sippola 2001
Filter efficiency (-)	0.098	0.49	0.74 (b)	0.88	Delp 2001

Note a: parameters ranged from 0.01 to 0.5 in sensitivity analysis
Note b: filtration efficiency ranged from 0 to 1.0 in sensitivity analysis

RESULTS AND DISCUSSION

Figure 3 shows, for the first four hours after the release, the fraction of anthrax in several representative storage locations. The airborne concentration in Room 4 rapidly declines within the first hour due largely to removal by deposition, by ventilation to outdoors, and by air exchange with other rooms. The response of Room 7 shown in Figure 3, typical of the response in the other five rooms, shows an initial increase in concentrations as air movement within the office suite transports contaminant from the source room. The moderate filtration efficiency used for the baseline case in this scenario limits, but does not prevent, the spread of anthrax via the HVAC system.

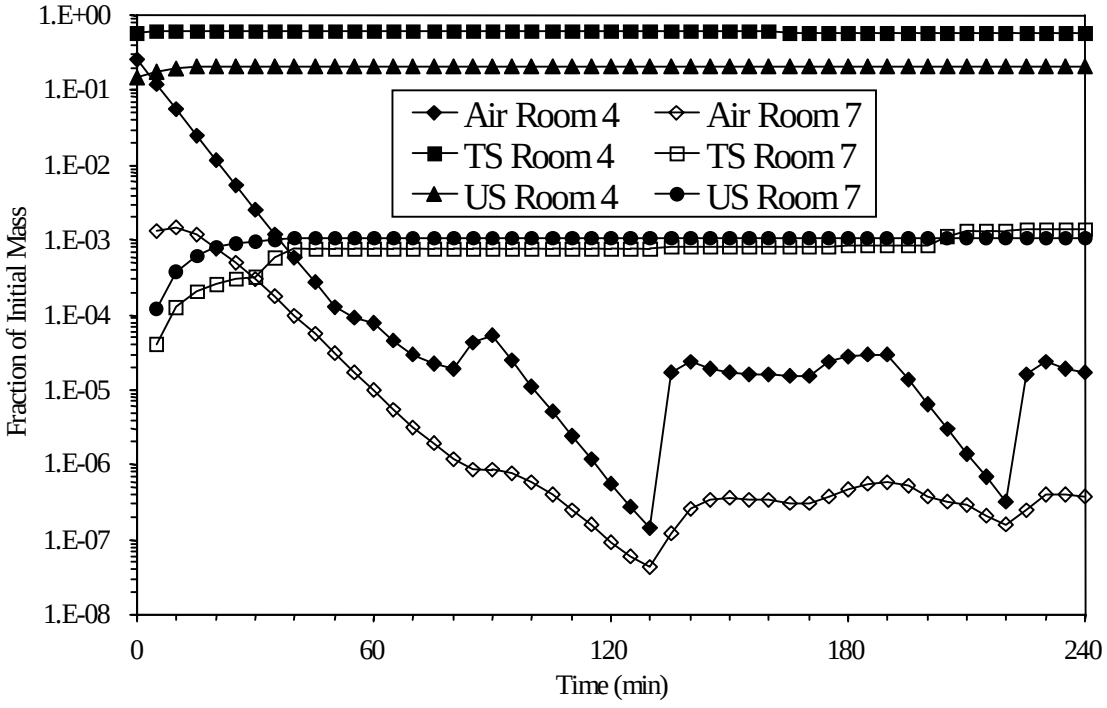

Figure 3. Mass fraction of anthrax in several representative storage locations as a function of time after release, where TS and US refer to tracked and untracked surfaces, respectively.

Although there is activity in both rooms, it is not until the airborne spore concentrations decline significantly that the effects of occupant activity can be discerned. For example, the concentration peaks seen at ~80 min result from resuspension. Furthermore, Figure 3 shows that occupant-related tracking increases the concentration on the tracked portion of the floor, relative to the untracked surface (the greater mass contained on the untracked surfaces reflects the fact that they take up a greater area than the tracked part of the floor).

These effects are also illustrated by the data in Table 2, which show a slight decrease in the mass fraction for all tracked surfaces over time, but an increase in the fraction of mass accumulating on the tracked surfaces when Room 4 is excluded. Simulations over greater time periods, or with more activity, would show even greater transport of spores due to human activity. The amount of mass collected on the HVAC filter, ~10%, reflects our choice of filtration efficiency. Note that under the simulation conditions we assumed, only ~ 6% of the anthrax is transported outdoors over the 48 hour time period.

Table 2. Fraction of released anthrax in various storage locations as a function of time

	Time (hr)					
Location	0.5	1	4	8	24	48
All tracked surfaces (TS)	0.62	0.61	0.61	0.61	0.61	0.61
All untracked surfaces (US)	0.21	0.22	0.22	0.22	0.22	0.22
All TS w/o Room 4	0.0066	0.0075	0.02	0.031	0.036	0.066
All US w/o Room 4	0.0082	0.0089	0.0089	0.0089	0.0089	0.009
Outside	0.059	0.060	0.060	0.060	0.060	0.061
HVAC filter	0.1	0.1	0.1	0.1	0.1	0.1
HVAC ductwork	0.0034	0.0035	0.0035	0.0035	0.0035	0.0035

The model calculates the integrated exposures for each of the six persons included in this scenario, based on the air concentrations in the rooms as functions of time and the activity pattern for each person. To convert the exposures to dose (in spores), we assume a constant breathing rate of 20 L/min and a bulk concentration for anthrax of 3×10^{10} spores per gram (Thatcher et al., 2000).

Table 3. Cumulative Doses (in spores) for selected time intervals for each day. The dose on day 2 is the incremental dose for that day.

		Time (hr)				
Person		0.5	1	4	8	10
3	day 1	1.0×10^5	1.1×10^5	1.1×10^5	1.1×10^5	1.1×10^5
	day 2	51	76	710	1900	2100
4	day 1	5.1×10^6	5.1×10^6	5.2×10^6	5.2×10^6	5.2×10^6
	day 2	2300	3300	1.1×10^4	1.7×10^4	2.0×10^4
5	day 1	2.2×10^5	2.4×10^5	2.4×10^5	2.4×10^5	2.4×10^5
	day 2	130	1200	2200	4500	5000
6	day 1	1.1×10^5	1.2×10^5	1.2×10^5	1.2×10^5	1.2×10^5
	day 2	53	80	330	530	790
7	day 1	1.1×10^5	1.2×10^5	1.2×10^5	1.2×10^5	1.2×10^5
	day 2	60	110	2300	3600	3900
8	day 1	0.0	0.0	190	420	420
	day 2	0.0	0.0	360	840	840

The resulting doses based on our scenario assumptions are shown in Table 3 accumulated for each 24-hour period. In this way, the additional doses obtained the second day can be seen more easily. The dose estimates in Table 3 can be compared with the dose at which 50 percent of an exposed population suffers lethal effects (the LD_{50}). For anthrax the LD_{50} is ~8,000 spores (Thatcher et al., 2000). Thus even very small fractions of the initial anthrax mass can pose a significant health risk - in some cases long after the initial release. Many of the second-day doses alone are close to the LD_{50}, while on the first day, the individual doses (excepting person 8) exceed the lethal dose by one to three orders of magnitude. On the other hand, for person 8, the cumulative dose on the second day is larger than for the first, due to the increased amount of anthrax accumulated on tracked surfaces, as shown in Table 2.

We examined the effects of two parameters, filtration efficiency and tracking, on exposures; parameter ranges are shown as footnotes to Table 1. Changing filtration efficiency illustrates the dispersion of anthrax within the office suite via the HVAC system. For some occupants, increased filtration reduces exposures dramatically in the first 24 hour period; however, this effect is not uniform, due to room-to-room airflow and tracking. For the second period, the changes in exposure are less significant, typically a 20 to 30% reduction. Changing the tracking parameter by a factor of 50 has a more modest effect on exposures. The largest effects, ~ 50% reduction in exposure, occur on the second day, when exposures are due to resuspension. Again this effect isn't uniform and depends upon the occupant activity pattern used in the scenario.

CONCLUSIONS

We developed a model to examine various scenarios for anthrax release and transport within buildings. These simulations help identify critical information or processes that contribute most to predicted outcomes or to uncertainties. This paper describes results from one of several scenarios and – based on this scenario – our simulations show the importance of airflow as a means of transporting aerosols through the building, with tracking having a more modest effect. However, activity-related resuspension is an important potential source of human exposure. Although advection and deposition are well understood, additional studies are necessary to understand better the effects of tracking and resuspension.

ACKNOWLEDGEMENTS

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MODELING PARTICLE DEPOSITION IN VENTILATION DUCTS

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ABSTRACT
This paper describes predictions from two models of fractional particle loss in four typical HVAC duct runs. One model is a state-of-the-art Eulerian formulation; the second is based on empirical fits to experimental particle deposition data collected in a laboratory. The experiments are briefly described and sample results are presented. The Eulerian model only predicts deposition from fully developed turbulence, while the empirical model can be applied to duct bends and developing turbulence as well. The models predict almost no losses for particles smaller than 1 µm and nearly complete loss of particles larger than 40 µm in all duct runs. The empirical model suggests that particle loss in ventilation ducts is dominated by gravitational settling to the floor of horizontal ducts, and by deposition to zones where turbulent flow is undeveloped, such as in bends and in duct sections immediately after bends.

INDEX TERMS
HVAC, particle deposition, ventilation ducts, modeling

INTRODUCTION
Particle deposition in heating, ventilating and air conditioning (HVAC) systems of buildings can affect system performance and influence the quality of indoor air. HVAC systems consist of louvers, filters, fans, heat exchangers, ducts and other equipment that provide conditioned air to interiors. Surfaces in ventilation systems can act as sinks, temporary storage reservoirs, and possibly sources, for pollutants such as particles, microorganisms and volatile organic compounds (VOCs).

Particles may deposit to and resuspend from duct surfaces. Predictions of exposure concentrations for building occupants are partially limited by an understanding of particle deposition in HVAC systems. Particle deposits have been observed to both sorb and emit VOCs to and from the passing air stream. Bacteria and fungi are known to deposit on HVAC surfaces and grow if sufficient water is present. Such growth produces VOCs and may amplify the concentration of microorganisms and certain VOCs in the air stream. Chemical interactions have been observed to occur between pollutants and HVAC surfaces (Morrison *et al*., 1998) and particle deposits may alter the nature of these surface interactions. These sorts of pollutant transformations may be of considerable importance in overall HVAC hygiene.

This paper applies two particle deposition models to ventilation duct runs to explore the degree to which particles are lost to duct surfaces as air travels from outdoors to the interiors of commercial buildings. The models give information on the expected location of particle deposits within HVAC ducts and can also be used to predict accumulation rates of particle deposits on duct surfaces. Experiments of particle deposition in a lab HVAC duct used for testing the Eulerian model and developing the empirical model are also briefly summarized.

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METHODS

Results of particle deposition experiments have historically been presented as plots of dimensionless deposition velocity, V_d^+ versus dimensionless relaxation time, τ^+. The dimensionless deposition velocity of a particle to a duct surface is defined as

$$V_d^+ = \frac{J}{C_{ave} u^*} \tag{1}$$

where J is the time-averaged particle flux to the surface, C_{ave} is the time-averaged airborne particle concentration and u^* is the friction velocity. The dimensionless deposition velocity depends on a variety of factors including particle size, air speed and the roughness of the deposition surface. The dimensionless relaxation time is defined as

$$\tau^+ = \frac{C_c \rho_p d_p^2 u^{*2}}{18 \mu \nu} \tag{2}$$

where C_c is the slip correction factor, ρ_p is the particle density, d_p is the particle diameter, μ is the dynamic viscosity of air, and υ is the kinematic viscosity of air. The penetration, P, of particles through a vertical duct section may be calculated if V_d^+ is known:

$$P = \frac{C_{outlet}}{C_{inlet}} = \exp\left(\frac{-4 L V_d^+ u^* P_{duct}}{U_{ave} A_{duct}} \right) \tag{3}$$

where C_{outlet} and C_{inlet} are the flow-weighted average particle concentrations at the outlet and inlet of the duct, respectively and L is the duct length, U_{ave} is the average velocity, A_{duct} is the duct cross sectional area and P_{duct} is the duct perimeter of a section through the duct normal to the flow direction. If the duct is oriented horizontally, differences in V_d^+ to the duct floor, wall and ceiling due to gravity must be taken into account and equation (3) is not valid. The fractional particle loss through a given section of duct is equal to 1 - P.

Experiments investigating the effect of air velocity and particle size on particle deposition rates from duct flow have been conducted in our laboratory. These experiments were carried out in a horizontal galvanized steel duct system with a 15×15 cm square cross section at air speeds of 2.2, 5.3 and 9.0 m/s and with monodisperse particles in the range 1-16 µm in diameter. Monodisperse, fluorescent particles were generated by a vibrating orifice aerosol generator (TSI Model 3450), neutralized by bipolar ions from a Kr-85 radioactive source (TSI Model 3054) and injected into the duct system through a mixing box. Fluorometric techniques were used to quantify the airborne particle concentration in the duct and the deposition flux to the galvanized steel duct surface. Particle deposition flux was measured at six locations for each experiment: two straight duct sections where turbulent flow was fully developed, two straight duct sections where turbulent flow was undeveloped and two 90° duct bends. The straight sections of duct where turbulent flow was fully developed were located at the end of long, straight sections of duct. The straight sections of duct where turbulent flow was undeveloped were located at the outlet of the mixing box and at the outlet of a 90° duct bend. In the straight duct sections, particle deposition fluxes to the upward facing duct floor, the vertical duct wall and the downward facing duct ceiling were measured independently to provide information on the location of particle deposits within the duct.

Data for particle deposition from fully developed turbulent flow generated by these experiments were compared to an Eulerian particle deposition model that accounts for Brownian and turbulent diffusion, turbophoresis, gravitational settling and surface roughness (Guha, 1997). This formulation represents the state-of-the-art in Eulerian deposition models and has shown good agreement with experiments conducted in small diameter tubes. This model can predict particle deposition to floor, wall and ceiling surfaces in ducts; however, it was developed to predict particle deposition only from fully developed turbulent flow.

Empirical correlations of the experimental data were developed to predict particle deposition in cases where the Eulerian model is not applicable or disagreed significantly with the current experimental data. In particular, the Eulerian model is not applicable in duct sections with developing turbulent flow or in duct bends. Furthermore, this model was found to disagree significantly with the experimental data for deposition to the duct wall and ceiling from fully developed turbulent flow. The only case where the Eulerian model agreed well with the experimental data was for particle deposition to the duct floor from fully developed turbulent flow. To address these limitations, we developed an empirical model that utilized our experimental data. An interpolation scheme was developed to enable the empirical correlations to be applied to the range of flow velocities and particle sizes of interest in ventilation ducts. The empirical model is in reasonable agreement with the limited data on particle deposition from flow in ducts with diameters similar to those found in HVAC systems. Furthermore, it allows for the calculation of particle deposition in duct bends and in straight sections of duct where turbulence is not fully developed. This is potentially important in HVAC systems where the turbulent flow is often not fully developed. However, this model is based on empirical fits to a limited set of experimental data and, as yet, has no means of accounting for the effect of duct surface roughness on particle deposition.

To explore the significance of particle deposition in typical HVAC ducts, these two particle deposition models were applied to the four duct runs described in Table 1. These duct runs were selected from a survey of 80 duct runs from four university buildings of 6- to 7-stories each. Duct runs were selected based on a random selection of duct endpoints and ranked in terms of length. In terms of total duct length, duct run A represents the 90^{th} percentile, duct runs B and C are at the 50^{th} percentile and duct run D corresponds to the 10^{th} percentile. Each duct run begins at the HVAC supply fan, ends at a supply register in the building and consists of a series of successively smaller ducts with decreasing flow rates from beginning to end. Airflow rates were assumed to be constant at the design rates and duct dimensions were obtained from design drawings. In all modeling, the ducts were assumed to be smooth.

Table 1. Description of modeled duct runs

Duct run	*Total length (m)*	*Horizontal length (m)*	*% Undeveloped turbulent flow*	*Number of bends*
A	72	51	57	8
B	48	15	50	3
C	48	35	58	7
D	28	20	65	6

RESULTS

Figure 1 shows an example of the experimental particle deposition data collected in a duct section where turbulent flow was fully developed. The left and right sides of this figure show the same experimental data collected at an air speed of 2.2 m/s ($u^* = 12$ cm/s) with 1, 3, 5, 9

and 16 µm diameter particles. The experimentally measured deposition rates to the duct floor are greater than deposition to the duct wall, which, in turn, are greater than deposition to the duct ceiling. Similar experimental results were observed at higher air speeds. On the left side of Figure 1, the Eulerian model agrees with the experimental data in the case of deposition to the duct floor, but predicts deposition rates to the duct wall and ceiling that are drastically less than observed experimentally. The right side of Figure 1 shows the empirical model fits to the experimental data.

Experimental results not shown here suggest that particle deposition rates in duct bends and in straight sections immediately after duct bends where turbulent flow is undeveloped are significantly enhanced compared to deposition rates in straight ducts with fully developed flow. The Eulerian model is not applicable to deposition in duct bends or in sections with undeveloped turbulence. Empirical correlations similar to those displayed on the right side of Figure 1 were developed for use in the empirical model to estimate particle deposition in bends and straight sections with undeveloped turbulence.

Figure 2 displays the fractional particle loss versus particle size for particles traveling through the four duct runs predicted by the Eulerian model. In this model application, particle deposition in duct bends was ignored and the enhancing effect of undeveloped turbulence after bends on deposition was also neglected. For all duct runs, particle loss is predicted to be negligible for particles smaller than 1 µm and nearly complete for particles larger than 40 µm. Duct run A, the longest duct run, is predicted to have the greatest losses for a given particle size in the range of 1-40 µm. The shortest duct run, however, is not predicted to have the lowest particle losses. Duct B run, the duct with the shortest horizontal length is predicted to have the lowest losses, partially because deposition to the floor of horizontal ducts is what controls particle losses when bends are ignored.

Further evidence that losses are controlled by deposition to the duct floor when deposition

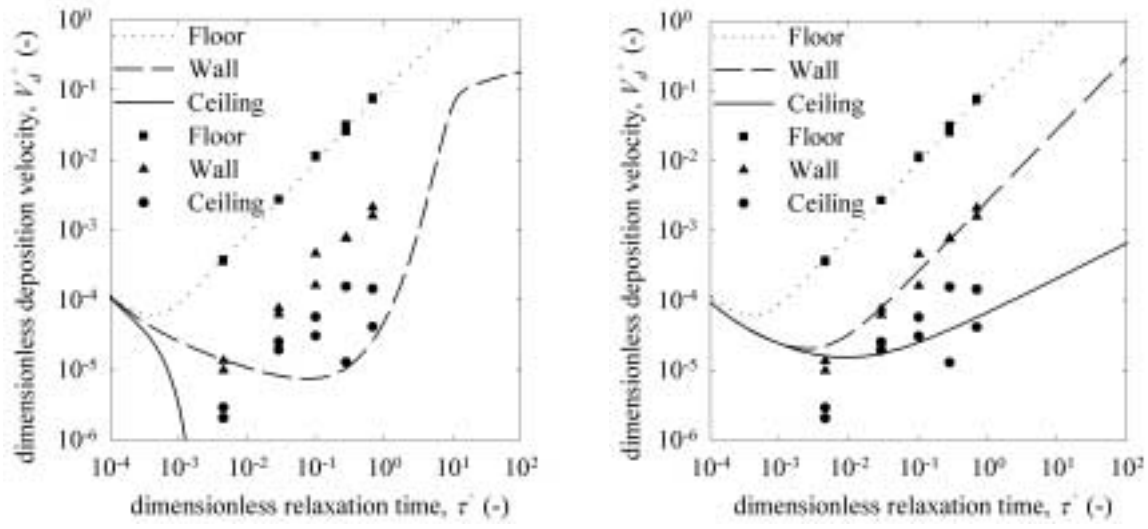

Figure 1. Comparison of the Eulerian model (left) and the empirical model (right) to experimental particle deposition data collected from fully developed turbulent flow at a velocity of 2.2 m/s.

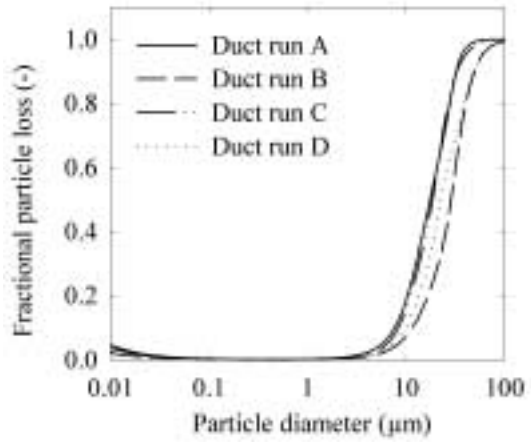

Figure 2. Fractional particle loss vs. d_p in modeled duct runs ignoring bends and flow within Eulerian model.

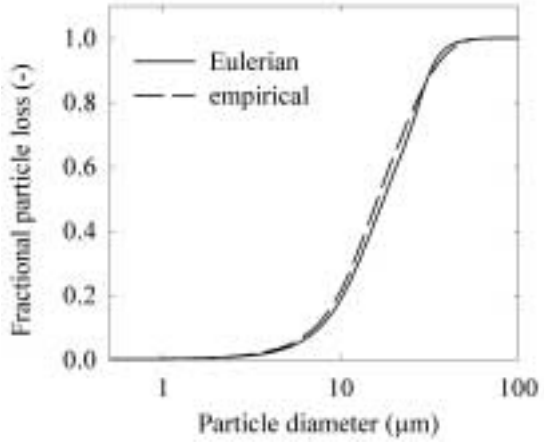

Figure 3. Fractional particle loss vs. d_p in duct run A ignoring bends and undeveloped undeveloped flow with both models.

in and after duct bends is ignored is found in Figure 3. Here, the Eulerian and empirical models predict virtually identical particle losses in duct run A even though the empirical model predicts much higher deposition rates to the duct walls and ceiling.

Figure 4 compares predictions by the empirical model for losses in duct run C when bends are ignored to the case where the effects of bends and the associated undeveloped turbulent flow are taken into consideration. In this figure, particle losses are observed to increase significantly for particles in the size range of 1-40 µm when bends are taken into account. Figure 5 displays predictions of particle losses in the four duct runs by the empirical model while accounting for deposition in bends and the enhanced deposition from the undeveloped turbulence after bends. In this case, particles larger than 20 µm are expected to be completely lost in duct runs A, C and D. Duct run B, with only three 90° bends, is predicted to have lower losses for 1-40 µm particles than the other duct runs which have at least 6 bends.

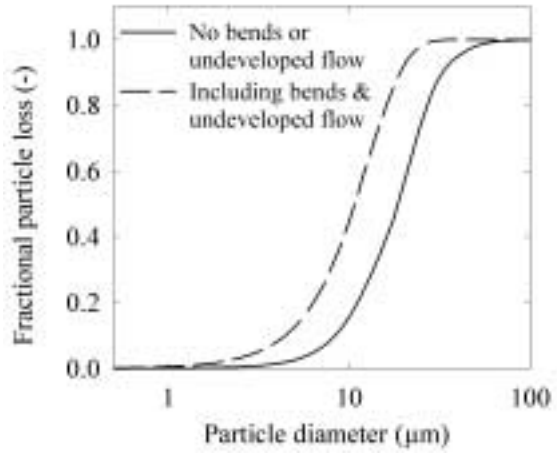

Figure 4. Difference in fractional particle loss in duct run C when including bends and undeveloped flow with empirical model.

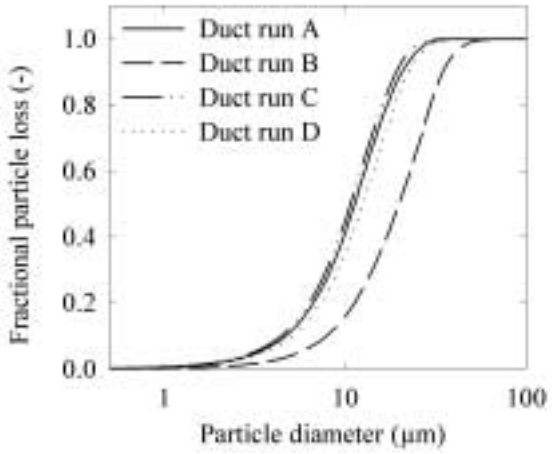

Figure 5. Fractional particle loss vs. d_p in modeled duct runs including bends and undeveloped flow with empirical model.

DISCUSSION

Experimental measurements suggest that, for particles larger than 1 µm, rates of deposition from fully developed turbulent flow to duct floors is significantly greater than to duct walls or ceilings over the range of air speeds of interest in HVAC systems. Experiments also indicate that particle deposition rates to duct surfaces are significantly enhanced in bends and in ducts immediately after bends compared to rates in duct with fully developed turbulence.

Even though the Eulerian and empirical models differ drastically in their estimates of particle deposition to duct walls and ceilings, predictions of fractional particle loss in sample ducts by the two models are nearly identical when bends are neglected. Both predict almost no particle loss in the modeled duct runs for particles smaller than 1 µm and complete loss for particles larger than 40 µm. Gravitational settling to the floor of horizontal ducts dominates particle losses in this case. When deposition in bends and enhanced deposition in sections after bends are considered using the empirical model, predicted particle loss rates for 1-40 µm particles are significantly increased compared to when bends are ignored. Our study results suggest that particle deposition in ducts primarily occurs in two zones: in and immediately after duct bends and to the floor of horizontal ducts.

CONCLUSIONS

A state-of-the-art Eulerian particle deposition model shows poor agreement with experimental data measuring deposition to the walls and ceiling of a horizontal ventilation duct. However, this poor agreement is inconsequential when predicting particle losses in straight HVAC duct runs because particle losses are dominated by gravitational deposition to the floor of horizontal sections, where the model performs well. The main deficiency of this Eulerian model in determining particle losses in HVAC ducts is its inability to account for deposition in bends and in straight duct sections with undeveloped turbulence.

The empirical model based on the collected experimental data is better able to deal with the bends and sections of undeveloped turbulence common to HVAC duct runs. Applications of this empirical model suggest that deposition in bends and in sections immediately after bends contributes significantly to fractional particle losses in typical HVAC ducts. Further experimental studies on particle deposition in duct bends and in sections following bends would help to reduce the uncertainties in this assertion. Additional experimental work is also warranted on particle deposition to rough duct surfaces, such as those covered with acoustic lining. The predictions that submicron particles are not significantly deposited in ducts should also be tested experimentally.

ACKNOWLEDGEMENTS

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RAPIDLY LOCATING SOURCES AND PREDICTING CONTAMINANT DISPERSION IN BUILDINGS

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ABSTRACT
Contaminant releases in or near a building can lead to significant human exposures unless prompt response measures are taken. However, selecting the proper response depends in part on knowing the source locations, the amounts released, and the dispersion characteristics of the pollutants. We present an approach that estimates this information in real time. It uses Bayesian statistics to interpret measurements from sensors placed in the building yielding best estimates and uncertainties for the release conditions, including the operating state of the building. Because the method is fast, it continuously updates the estimates as measurements stream in from the sensors. We show preliminary results for characterizing a gas release in a three-floor, multi-room building at the Dugway Proving Grounds, Utah, USA.

INDEX TERMS
Multizone modeling, Bayesian statistics, COMIS, Parameter estimation, Sensors

INTRODUCTION
Effective response to unexpected pollutant releases in buildings often requires knowing the source locations, the amounts released, and the duration of the event. However, merely measuring airborne pollutant concentrations using sensors may not reveal this information. Complex airflows typically found in multi-room, multi-floor buildings will often quickly disperse the pollutant throughout the building, leaving insufficient time for the sensors to adequately sample the temporal and spatial profiles of the event. Therefore, sensor measurements must first be interpreted. Moreover, they must be interpreted quickly and continuously as the measurements stream in from the sensors.

Traditional data interpretation algorithms, like optimization, Gibbs sampling, and Kalman filtering, are inadequate for this task. They rely either on simplifying assumptions that are not often met in many indoor pollutant transport systems, or on time-consuming inverse models, which must repeatedly run computationally-intensive fate and transport models after an event has begun (Sohn et al., 2002).

We present an alternative algorithm which uses Bayesian statistics. Our approach succeeds, where traditional methods fail, because it decouples the simulation of predictive fate and transport models from the interpretation of measurements. Time-consuming airflow and pollutant transport predictions and uncertainty estimates are computed prior to a pollutant release. This allows for rapid data interpretation during an event. The technique may be used to estimate the location, magnitude, and duration of the release, to characterize any unknown or variable building or weather conditions, and to predict future pollutant transport in the building. Though Bayesian statistics have been applied to several environmental fields (see

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Sohn et al. 2002 for a list of recent applications), using it to decouple indoor airflow and transport modeling from data interpretation has, to our knowledge, not been previously reported in the literature.

The objectives of this paper are thus to (1) present our Bayesian approach for interpreting sensor data in real time, and (2) demonstrate the approach by successfully detecting and characterizing a pollutant release in a real multi-floor building.

APPROACH

The Bayesian data interpretation approach is divided into two stages. First, in the *pre-event* or *simulation* stage, all of the time-consuming tasks associated with data interpretation are completed before a pollutant release occurs. A model of the building's indoor airflow and pollutant transport is developed, and input parameters for the model are selected. Any unknown or variable model input, like the location and duration of the pollutant, or the HVAC operating mode, is assigned an uncertainty distribution that describes the probabilistic range of possible values. Generally, wide distributions are assigned given the limited prior information. Next the user generates a library of model realizations by sampling the space of the model parameters using Monte Carlo, or other sampling technique, and predicting airflow and pollutant transport for each set of parameters. Each model realization and model simulation represents a possible building operating condition and pollutant release scenario. Thus, sufficient sampling of the uncertainty distributions is essential to represent the full range of possible building and pollutant release conditions. The resulting library of simulations may consist of several thousand scenarios.

The second stage of the Bayesian approach, *data interpretation*, takes place during a release event. The algorithm compares data streaming in from sensors to the library of pollutant transport predictions using a structured probabilistic method referred to as Bayesian updating (Brand and Small, 1995 and Sohn et al. 2002). Bayes' rule allows us to quickly estimate, and update, the level of agreement between model predictions and the observed data while accounting for the effects of error in the measurements, correlation or averaging of the spatial and temporal data, and any imperfect model representation. See Sohn et al. (2002) for a full description of the technique. To summarize the process, each realization in the library is compared to the data to assess the likelihood that the realization describes the event in progress. A realization with predictions that fit the sensor data well will have a high likelihood estimate. This in turn suggests that the model inputs used to generate that realization in the pre-event simulation stage has high probability of describing the event in progress. Comparing the relative fits for each realization using Bayesian statistics allows us to estimate the best-fitting suite of model inputs and the associated uncertainty.

This second stage of the approach is mathematically simple and can be executed very quickly, much quicker than the rate at which new data will likely arrive from sensors. As long as the original library of simulations provides adequate coverage of the model and input parameter space, the data interpretation during the event can be conducted without further evaluation of the flow and transport models.

APPLICATION

We applied our approach to locate and characterize a pollutant release in a three-floor, multi-room building at the Dugway Proving Grounds, UT. Figure 1 illustrates the building and shows the floorplan of the first floor. The first and second floors each consist of three rooms

and a stairwell landing. The third floor consists of a large attic space and a stairwell landing. An air handling unit (AHU) supplies air to the first and second floors and returns air only from the first floor. Researchers from Lawrence Berkeley National Laboratory (LBNL) conducted extensive blower door tests on the building to determine interzonal flow parameters and leakage rates. They also conducted twelve tracer experiments in the building releasing puffs of propylene gas at various release points and under several operating conditions of the AHU. Details of the experimental design were discussed by Sextro et al. (1999).

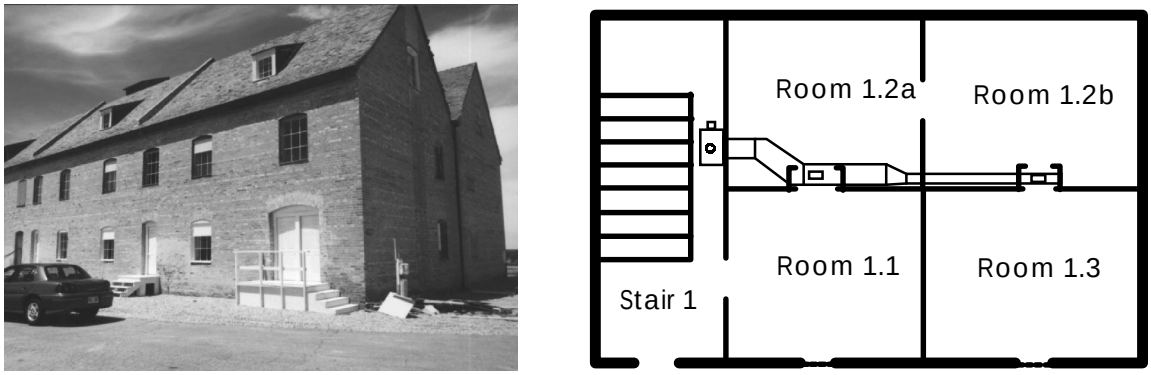

Figure 1: The three-floor building and plan view of the first floor with air handling unit.

Table 1. Uncertainty in the source and building characteristics.

Parameter	*Range*
Source Location	10 locations, consisting of any of the rooms and stairwell. Each location is assumed to be equally likely.
Source Duration	1 sec to 5 min. Log-uniform distribution.
Source Amount	10 to 100 grams. Log-uniform distribution.
Status of Door Positions	3 scenarios, all equally likely: (1) all interior doors open, (2) all interior doors closed, (3) stairwell doors closed, all others open.

In the simulation stage of the Bayesian approach, we hypothesized about the types of pollutant releases that might occur in the building, and assigned uncertainty ranges to the release characteristics (Table 1). We then generated a library of five thousand airflow and pollutant release scenarios by sampling the uncertainty ranges using Latin Hypercube sampling techniques. Airflow and pollutant concentrations in air were predicted for each scenario using the COMIS model, which had been validated previously with experiments conducted in this building (Sextro et al. 1999).

In the data interpretation stage, sensors would be placed at various locations in the building to measure airborne concentrations. The sensors would report the measurements in real-time to the monitoring computer, where they would be interpreted by the algorithm. In the actual experiments conducted at Dugway, propylene detectors were placed in every room and at each stair landing, for a total of eleven sensors. Concentration data were recorded continuously at 20 second intervals. A sample of the data from one of the experiments is shown in Figure 2 at

three locations within the building for the first ten minutes of the experiment. In order to test our algorithm, we used data from this experiment, which consisted of a release of ~20 grams of propylene gas into the HVAC return intake in room 1.2a with the HVAC operating. To ensure that our test was valid, we 'blinded' our interpretation algorithm to the actual location and release conditions (see Table 1). The interpretation method was applied for an unknown source location, just as would be the case for a potential release in a real building. For our simulated exercise, we assumed that each data point was reported to the monitoring computer – in this case essentially simultaneously, though the reporting could be asynchronous.

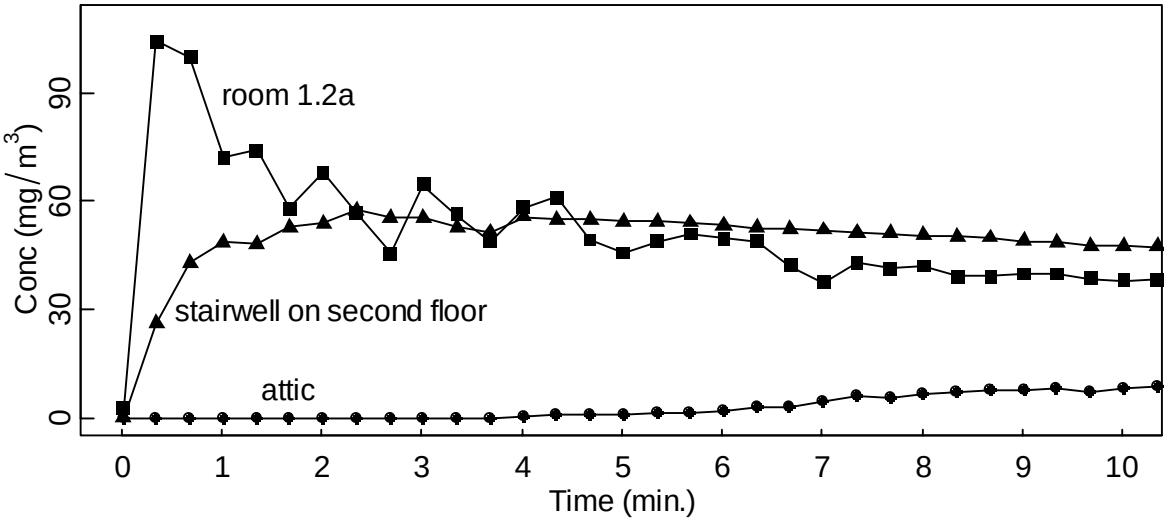

Figure 2: Sensor measurements in room 1.2a (first floor), the stairwell landing on the second floor, and the attic (third floor). Twenty grams of propylene were released in room 1.2a in one second.

Based on the data from all eleven sensors, the interpretation algorithm reports estimates for all of the unknown model inputs, which are updated every 20 seconds. Figures 3a and 3b show the source location probability for two different rooms. It should be noted that at time t=0, the probability of the source location is equal for each room. As can be seen in Figures 3a and 3b, the interpretation algorithm takes less than two minutes to identify the source location (room 1.2a) with high probability.

Since sensors were placed in each room, one might conclude that the location with the highest concentration would always be the release location However, that is correct only when the air is sampled when the source is on and releasing at a constant rate. In this experiment, the release lasted for only one second and the sensors first sampled nineteen seconds after the source stopped. The estimation is further complicated by the AHU quickly dispersing the pollutant throughout the building.

As an example of updating other uncertain model input parameters, Figure 4 shows the algorithm's estimate of the total mass released as it refines that estimate every 20 seconds. The median estimate quickly converges on the correct amount released and uncertainty gradually narrows with each sampling interval. Similar results were found for the other unknowns identified in Table 1, but are not shown.

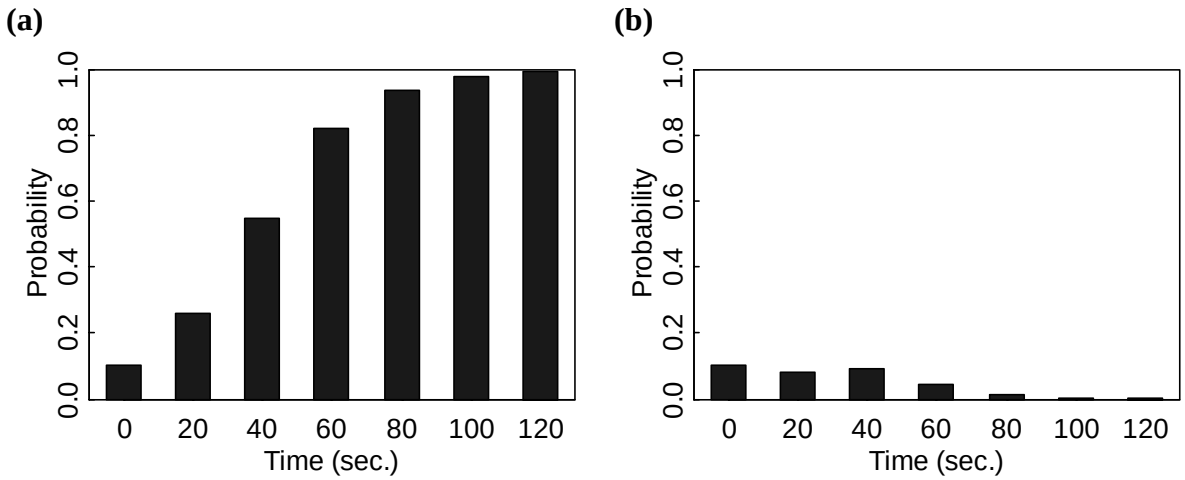

Figure 3: Estimates of the probability that the source is located in room (a) 1.2a and (b) 2.2. At t=0 seconds (before data interpretation begins), the source is assumed be in any of the ten locations with equal probability (i.e., probability = 1/10).

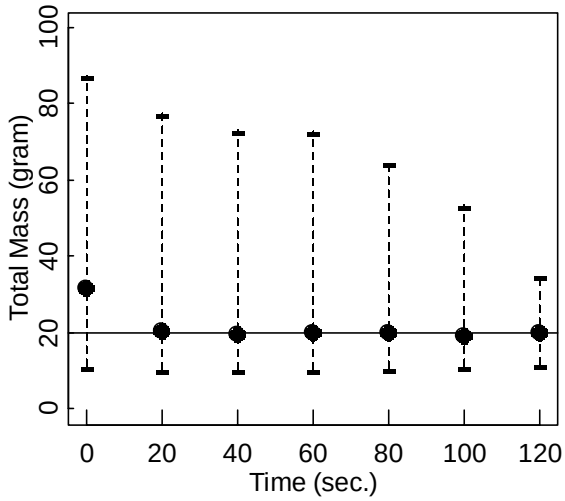

Figure 4: Estimates of the total amount released. At t=0 seconds (before data interpretation begins), the estimate is based on the initial uncertainty defined in Table 1. The solid circle and uncertainty range represent the median and 90 percent confidence interval, respectively, and the horizontal line denotes the actual amount released.

CONCLUSION AND IMPLICATIONS

This paper presents a Bayesian approach for interpreting sensor measurements in real time. It differs from other model parameter estimation methods by decoupling the simulation of airflow and pollutant transport from the interpretation of measurements. This allows us to divide the data interpretation into two parts. The simulation stage completes all of the time consuming tasks, such as development of airflow and pollutant transport models, uncertainty characterization, and simulation of pollutant transport, and compiles the scenario simulations

into a library of results. The data interpretation stage quickly accesses this library as data stream in during an event.

We demonstrated the approach by analyzing a gas release in a three-floor building. While the results are preliminary, they illustrate how our approach can quickly interpret data. The approach quickly and correctly identified the source location and the release amount. Though not illustrated here, the algorithm also correctly identified, in less than two minutes, both the duration of the tracer gas release and whether doors were open or closed. Lastly, it correctly predicted the future dispersion of the pollutant in the building.

In future work, we will use our approach to guide sensor deployment. Decoupling data interpretation from model evaluation allows us to compare the performance of many hypothetical sensor operating conditions and sensor locations. Such comparisons could help identify the requirements for a sensor network, including the number, sensitivity, and response time of sensors, based on the desired performance of a data interpretation algorithm in any given building.

ACKNOWLEDGEMENTS

This work was supported by the Office of Non-proliferation and National Security, Chemical and Biological Non-proliferation Program of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098. P. Reynolds was supported in part by the U.S.DOE Energy Research Undergraduate Laboratory Fellowship Program.

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III. ENVIRONMENTAL TOBACCO SMOKE

A PILOT STUDY OF THE BEHAVIOR OF GAS- AND PARTICLE-PHASE ETS TRACERS IN RESIDENCES

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ABSTRACT

Our previous study of environmental tobacco smoke (ETS) in a three-room environmental chamber showed that smoking history significantly influenced inter-room ETS transport , particularly of gas-phase nicotine. We conducted a three-home pilot study where smoking was limited to one room. Single-smoker residences were monitored during five one-week periods while the smoker participated in a smoking cessation program. Nicotine traced ETS particles were detected reliably in the smoking rooms (SRs) and unreliably in the non-smoking rooms (NSRs). On average, the ventilation- and volume-normalized smoking rate, 0.1 Cigarette-$h^{-1}m^{-3}$, added about 17 and 4 μg $m^{-3}$of ETS particles into the SR and NSR, while average nicotine concentration increases were 2 and 0.06 μg m^{-3}, respectively. Thus, nicotine tracers may underestimate ETS particle exposure in a NSR (e.g., a child's bedroom) by a factor of 2 to 8. In other words, ETS exposure predicted from nicotine concentrations could be almost an order of magnitude lower than actual exposure.

INDEX TERMS: Environmental tobacco smoke, Exposure assessment, Field study, FPM, Nicotine, Residence, Sorption, UVPM

INTRODUCTION

ETS is a significant contributor to the concentrations of respirable suspended particles (RSP) in indoor environments where smoking occurs (Spengler *et al*, 1981; NRC 1986). ETS exposure assessment in indoor environments has been based primarily on measurements of gas- and particle-phase chemical tracers. Our previous studies of ETS (Apte *et al*., 1999 and 2002) indicated that inter-room mixing in a three-room environmental chamber affected individual ETS constituents differently, depending upon both the chemical identity of the ETS constituent and its phase (gas or particle). Nicotine from ETS in the smoking room was virtually undetected in the corridor or the non-smoking room, even when ETS particles were easily detected, and even when the door openings allowed for free air exchange between the rooms. The rate of sorption onto and into chamber surfaces dominated the mass transfer dynamics.

These results also suggested that ultraviolet absorbing particulate matter (UVPM) and fluorescent particulate matter (FPM) could trace ETS particles more accurately than nicotine when measurements are made in environments with varying or unknown conditioning of interior surfaces (smoking history). ETS in the SR was used to calibrate UVPM so that the

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ratio of UVPM to respirable suspended particles (RSP) averaged 1.0 in the smoking room. That ratio was 0.8 in the corridor and nonsmoking room, indicating a possible 20% reduction of PM tracer specificity through transport and loss mechanisms such as selective volatilization of UV-absorbing material from the particles. In contrast, the ratio of nicotine to RSP was about 0.2 in the smoking room, and dropped to about 0.05 in both the corridor and NSR. This 400% reduction in the nicotine to RSP ratio suggests a potential for significant underestimation of particle-phase ETS exposure when nicotine is measured in non- or lightly-ETS-conditioned environments that connect to rooms where smoking occurs.

Thus, the reliability of ETS tracers in actual home or building environments should be examined in light the following criteria for tracer effectiveness:

1. Tracers must be conservative, remaining in constant ratio to the ETS constituent of interest. Exposures to ETS aerosols are generally regarded as the source of the respriratory health effects; hence tracers must reflect changes in ETS particle concentration as the ETS dilutes, ages, and moves between rooms, even in the presence of other sources of particles. Conservative tracers must also be chemically stable over the time of both collection and analysis.

2. The analytical methods for tracers must have adequate sensitivity and selectivity.

3. The analytical methods must be cost effective. This is especially important for large-scale exposure studies where many samples will be processed. The main issue is balancing precision and accuracy with analysis cost.

A three-home pilot field study was conducted to: (a) examine the reliability of various ETS particle tracers in real indoor environments; (b) estimate potential bias of nicotine-based exposure assessments due to nicotine sorption and re-emission; (c) estimate potential bias of particle-mass based exposure assessments due to interference from other particle sources; and (d) quantify non-smoker ETS residential exposure reduction due to smoking cessation.

Table 1. Instrumentation and measurement methods for 3 home ETS pilot field study.

	Sampling Method	*Instrumentation*	*Analytical Method*
Nicotine	Passive diffusion, treated filter	Hammond Passive Sampler (see Hammond and Leaderer, 1987)	GC-NPD
RSP (PM 3.5)	Gravimetric air sampling at 1.7 L· min^{-1}	10mm Nylon Cyclone, Teflon Coated Glass Fiber Filter	Gravimetry
UVPM,FPM	As PM 3.5	Analysis of PM 3.5 filter	HPLC
Ventilation Rate	Peristaltic pump, Tedlar source and sample collection bags	Continuous injection and sampling of sulfur hexafluoride (SF_6)	GC-ECD

METHODS

Participants were recruited from members of a six-week smoking cessation class conducted by the Department of Health and Human Services, Tobacco Prevention Program, Berkeley, CA, USA. Each participant was the only smoker in the household, and smoking always took place at the same location in the house. Weekly smoking rate was monitored by cigarette butt

count. Houses were monitored for ETS and nicotine during weeks 1, 2, 3, 4 and 6, and the smoking cessation class encouraged abstinence beginning with week 3.
The five one-week integrated measurements included nicotine, particle mass, UVPM, FPM and time averaged whole-house ventilation rate (Table 1). RSP was sampled with a 10 mm nylon cyclone (50% cutpoint = 3.5 μm, flowrate 1.7 L min^{-1}) and collected on pre-weighed, pre-cleaned filters. The homes were visited weekly to recover the samples and deploy new sampling media.

Particle and nicotine samples were collected indoors in the rooms identified as the main smoking room and the non-smoking room. Outdoor PM 3.5 was also measured weekly. These outdoor samples were used to adjust for any infiltrating particles or particle-bound tracers. Particle mass was determined gravimetrically. UVPM and FPM concentrations were analyzed from extracts of the PM 3.5 particle filter samples (Apte *et al*., 2002). Solanesol analyses were conducted, but solanesol was detected in only one location, the SR used by the heaviest smoker. We suspect that low concentrations of solanesol were not stable over the one-week sampling period.

The limit of detection (LOD) of the nicotine, based on field blanks, is about 0.07 μg m^{-3}. Likewise, uncertainty based on duplicate measurements is about ±10% for nicotine greater than 0.15 μg m^{-3} (LOD*2) and ±50% for those less than 0.15 μg m^{-3}. Estimated UVPM and FPM uncertainty ranged from ±30% to ±50%, and from ±40% to ±80%, respectively, depending on the concentration range (Apte *et al*., 2002). Uncertainty in the PM 3.5 measurements was less than ±10%.

Whole-house air exchange rate was measured by decay of sulfur hexafluoride injection (SF_6). An SF_6 injection suitcase was located in a central (non-smoking) room about 1-2 m above the floor. Two sampling suitcases were used, one in the smoking room and one in the non-smoking room. Weekly-average indoor SF_6 concentrations ranged from 18 to 780 μg m^{-3} during the study.

Although the exact smoking time and emission rate profile would be needed to calculate the true week-average ETS concentrations, the steady-state mass balance model can be used to compare, on an equivalent basis, the measured ETS particle and particle tracer concentrations across study weeks and between houses. The steady-state form of the mass balance equation (Traynor et al., 1989) is:

$$C_{ss} = \frac{Pa\,C_o + S/V}{(a+k)} \qquad (1)$$

where
C_{ss} = the steady-state particle or particle tracer concentration (μg m^{-3}),
P = penetration fraction of particles from outdoors (0-1, unitless)
C_o = outdoor concentration of particles or particle tracers (μg m^{-3}),
S = the ETS particle or particle tracer source strength (μg h^{-1}),
V = the house volume (m^3),
a = the whole-house air exchange rate (h^{-1}), and
k = the particle deposition decay rate (h^{-1}).

Weekly cigarette butt count was used as a surrogate for S, assuming that this count was proportional to the weekly average ETS particle emission rate. Values of 0.08 h^{-1} and 0.7 were assumed for k and P, respectively (Traynor et al., 1989). We have used a value for k that

represents particle sizes typical of ETS. Measured house volume and weekly average air exchange rate values were used for *V* and *a*, respectively. The results for indoor PM 3.5 and ETS tracers have been adjusted by subtracting the contribution of outdoor air, $PaC_o/(a+k)$. With the assistance of the study participants we identified rooms in each home in which smoking did or did not occur and designated them as the SR and NSR, respectively. In House 1, a single story, single family residence, the SR was the kitchen. House 2 was a second-story flat of a large 2-floor wooden structure, and the SR was the living room. House 3 was also the second story of a small two-story wooden structure and the SR was the smoker's bedroom.

RESULTS

Figure 1 shows the measured weekly SR and NSR PM3.5 concentrations (adjusted for infiltrating outdoor PM 3.5) for the three study households. The clear downward trends in adjusted indoor PM 3.5 during the course of the smoking intervention reflect the success of the participants in reducing their smoking rate.

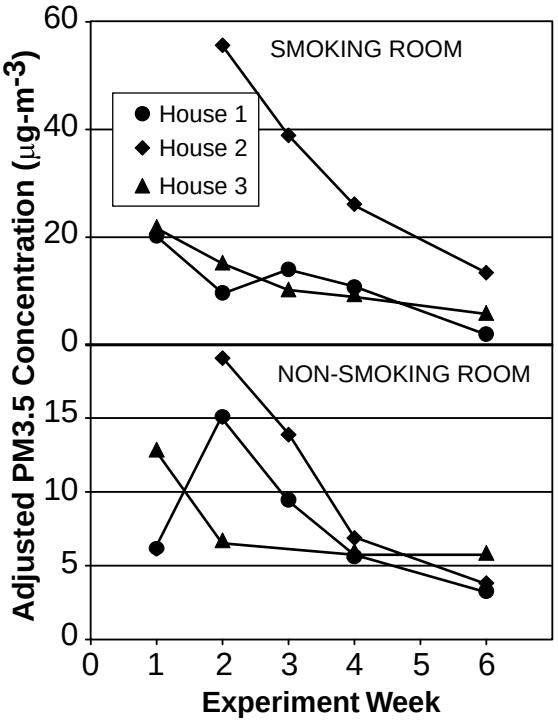

Figure 1. Adjusted weekly PM 3.5 concentrations in three study homes.

Figures 2 and 3 present the nicotine and ambient-adjusted PM 3.5, UVPM, and FPM indoor concentrations for the SR and NSR, respectively. Based upon the limited data from 3 houses, these figures should be considered suggestive of ETS behavior, but not conclusive. The ETS tracer concentrations are plotted against the household ETS source strength calculated from the weekly butt-count and ventilation rate data. Presenting the data this way allows examination of tracer behavior across the three houses. All the tracers performed similarly in predicting weekly smoking rates (and thus ETS emission rates) in the SRs in real homes – correlation between butt-count based source strengths and all tracers have R^2 values from 0.6 to 0.7.

For the NSRs the relationship between the tracer concentrations and the ETS source strength is much less clear. In Figure 3, a week 1 nicotine measurement in the NSR (0.003 μg m^{-3}) was removed from the figure, as it was an extreme outlier and possibly suspect. If this data point were included, nicotine would have no predictive value (R^2 =0.03, slope=0.70). Nicotine and FPM had poor predictive ability when NSR data from all houses were considered together. UVPM, with an R^2 of 0.5, did the best at predicting NSR ETS concentrations, while adjusted PM 3.5 had much lower R^2 values, possibly due to other indoor sources of respirable particles. Overall, none of the ETS tracers provided particularly strong predictive power for the ETS source in the NSR. Even so, UVPM and FPM, when adjusted for infiltration of outdoor PM, did meet the criteria (given in the introduction) for tracer effectiveness in both the smoking and non-smoking rooms.

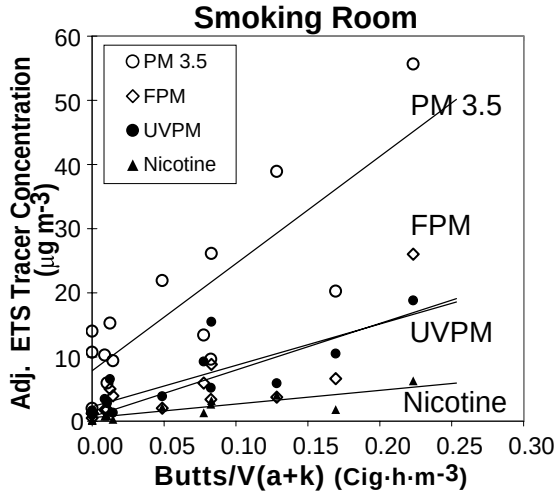

Figure 2. Adjusted ETS tracer concentrations in the SR vs. butt-count based source strength for all three study homes. Slope (R^2) for regression fits are 167 (0.68), 73 (0.63), 64 (0.68), and 21 (0.73) for PM3.5, FPM, UVPM, and nicotine, respectively.

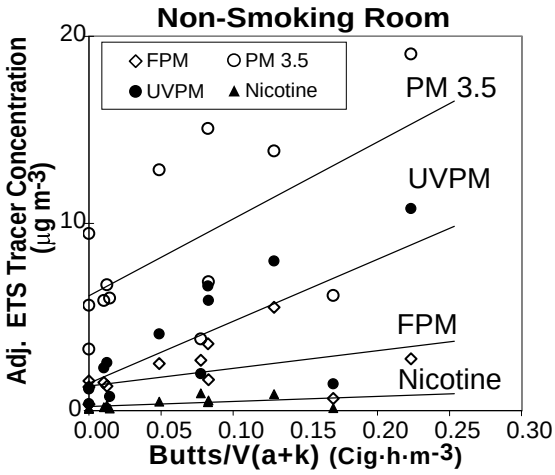

Figure 3. Adjusted ETS tracer concentrations in the NSR vs. butt-count based source strength for all three study homes. Slope (R^2) for regression fits are 41 (0.37), 33 (0.54), 9 (0.20), and 2.7 (0.26) for PM3.5, UVPM, FPM, and nicotine, respectively.

Interestingly, after adjustment for infiltrated particles, PM 3.5 rather than UVPM or FPM, provided the most consistent and highest correlation with ETS source strength. This is not likely to occur in homes with significant indoor sources of RSP such as combustion-generated particles from fireplaces and candles.

Nicotine traced ETS well in the smoking areas of all three houses, but was inconsistent in the non-smoking rooms where surfaces were probably not saturated with nicotine. The variability in nicotine concentrations in the NSRs could be attributed to the absence of information about the rates of sorption/desorption processes and inter-room air transport. In contrast, the PM tracers did follow ETS reliably in areas with unknown smoking histories and inter-room transport rates.

DISCUSSION AND CONCLUSIONS

In this study, for every 0.1 Cig-h^{-1}-m^{-3} of smoking about 17 μg m^3 of elevated ETS RSP could be expected in the SR, and 4 μg m^{-3} in the NSR. Similarly, predicted increases in nicotine concentrations at the same normalized smoking rate would be 21 μg m^{-3} and 0.3 μg m^{-3}, respectively. Thus, in these houses the concentration of ETS particulate matter in NSRs was about 25% of that in the smoking room, while on average, only about 13% of the nicotine was transported. Based on a detailed examination of the pilot study data, we conclude that an estimate of ETS particle exposure based on nicotine measurements in the NSR might be biased low by a factor ranging from two, to eight if the low nicotine value is included.

The pilot field study did not uncover significant bias from other ambient particle sources during spring weather in residential neighborhoods. Evaluation of the selectivity of UVPM for ETS compared to other particle sources requires supplementary measurements of ETS in the presence of wood smoke and diesel exhaust (Gundel *et al,* 2000). Our initial assumption was that indoor respirable particle sources such as cooking or vacuuming would be negligible compared to ETS. However, we found that indoor sources of respirable particles without a UV absorbing or fluorescing component may have been present in concentrations similar to the ETS particles. Certainly, as the ETS concentrations declined with the residents' smoking

cessation efforts, other indoor sources would become more dominant sources of particles. Additionally, the actual smoking rates in two houses were lower than expected.
Based on this pilot study, indoor PM 3.5, UVPM, and FPM and nicotine correlated well with ETS emission rates (calculated from cigarette butt counts and ventilation measurements as discussed above) in household smoking rooms. UVPM tracked reliably even in non-smoking rooms after all three smokers reduced their smoking rate by at least half during the six-week program. Additional method development is necessary to reduce the uncertainty in PM tracer quantitation. Furthermore, a larger study sample size would be necessary to make any clear conclusions and to validate our preliminary finding that exposure estimation based on nicotine measurements can be complicated and potentially biased.

ACKNOWLEDGEMENTS
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CHARACTERIZING SIZE-SPECIFIC ETS PARTICLE EMISSIONS

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ABSTRACT
We report a method for estimating the size distribution of particle emissions from indoor sources. The method is applied to concentration data from a series of cigar and cigarette experiments to characterize environmental tobacco smoke particles. The method incorporates a particle dynamics model, which provided good fits to observed concentrations when using, as input, optimal values of mass emission rate and deposition velocity for each particle size range. The optimal particle emissions were fit to log-normal distributions, yielding mass median diameters of approximately 0.2 μm and an average geometric standard deviation of 2.3. The total particle emissions obtained by integrating the empirical size distribution were 0.2 - 0.7 mg/min for cigars and 0.7 - 0.9 mg/min for cigarettes. The measurements of particle size characteristics agree well with prior research, but the integrated mass measurements are consistently lower than those determined from filter-based measurements.

INDEX TERMS
Environmental tobacco smoke, Particle emissions, Particle size distribution, Modeling, Sources

INTRODUCTION
Accurate modeling of environmental tobacco smoke (ETS) particle concentrations can improve our understanding of human exposure to ETS particles, including the regional deposition of ETS particles in the lung. To this end, we seek an improved characterization of the particle size distribution of ETS emissions, as well as an evaluation of model performance and a better understanding of ETS particle dynamics.

Because size is a major factor controlling airborne particle behavior, many previous investigators have characterized the particle size distribution of mainstream, sidestream, or ETS emissions (Chung and Dunn-Rankin, 1996; and Chang et al., 1985), including aged and diluted sidestream smoke (Ingebrethsen and Sears, 1989) and smoke generated by a person in a room-sized chamber (Kleeman et al., 1999). The particles in ETS, having undergone mixing and dilution over varying time scales, have size distributions that can be quite different from the distributions of fresh mainstream or sidestream smoke. While undergoing dispersion, particle mass can evaporate (Hinds, 1978), leading to smaller-sized particles, or coagulate, leading to larger-sized particles. Particle-size distributions may also be transformed by particle deposition on room surfaces and removal by filters.

The approach we follow in the present work is based on a preliminary effort by Sextro et al. (1991), and consists of the analysis of measured time-dependent ETS concentrations using a particle dynamics model developed by Nazaroff and Cass (1989). The model is fitted to the

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experimental data with emissions and deposition profiles as unknown inputs. By adjusting the input parameters such that an optimized fit is obtained, important information can be obtained about ETS particles. The quality of the fits gives an indication of model accuracy.

METHODS

For this study, we used data from experiments by Klepeis et al. (1999) in which a single cigarette or cigar was machine-smoked inside a low-ventilation stainless steel chamber. In these experiments, ETS particle concentrations were measured in 1-minute intervals using an optical particle counter (PMS-LASAIR; 0.1 - 2 μm diameters) and over 10- or 30-minute scans using a differential mobility particle sizer (TSI-DMPS; 0.01 - 1 μm diameters). Total particle mass (TPM) concentrations were also measured using Teflon filters, which were weighed before and after each experiment.

We adapted the aerosol dynamics model of Nazaroff and Cass (1989) to calculate ETS-particle concentrations for each experiment, taking into account the effects of ventilation, particle coagulation, particle deposition, and direct particle emissions from a cigar or cigarette. The model does not incorporate particle evaporation or condensation, although Kousaka et al. (1982) report that humidity only alters the size distribution of tobacco smoke particles under supersaturated conditions, which do not apply in our case. Ventilation was measured from tracer gas decay, and an ETS particle density of 1.1 g/cm^3 (Lipowicz 1988) was used for conversion between number and mass concentration. The unknown model parameters, which we intend to characterize, are size-specific particle deposition (quantified as a deposition velocity) and size-specific mass emission rate.

To determine the unknown parameters for each of the measured particle size ranges (or *bins*), our task is to find the mass emission rate and deposition velocity for each bin that result in the best fit of model predictions to measured concentrations. In mathematical terms, we are solving an *inverse problem* (Rabitz and Alis, 2000). The fits were accomplished using a search method where values for emissions and deposition velocity were varied in a grid pattern until the minimum mean absolute deviation between the modeled and observed time series was encountered (see Figure 1). We term this procedure *parameter optimization*, since we are concerned with finding *optimal* values for model input parameters (see Law and Kelton, 1991, Chapter 12). Initial guesses for each optimization were determined from the observed peak concentrations and decay rates for each size bin (using the LASAIR data), assuming independence between the bins. Initial guesses for particle sizes below 0.1 μm, which were missed by the LASAIR, were obtained from the DMPS data and then optimized against all of the larger LASAIR bins.

After the optimization procedure was completed for each experiment, we fit a log-normal distribution to the mass emission rate to obtain estimates for the mass median diameter (MMD) and geometric standard deviation (GSD) of the emissions size distribution. These fitted parameters are identical for the distributions of total mass emissions (mg), emission rate (mg/min), and mass-normalized emissions (mg/g-smoked).

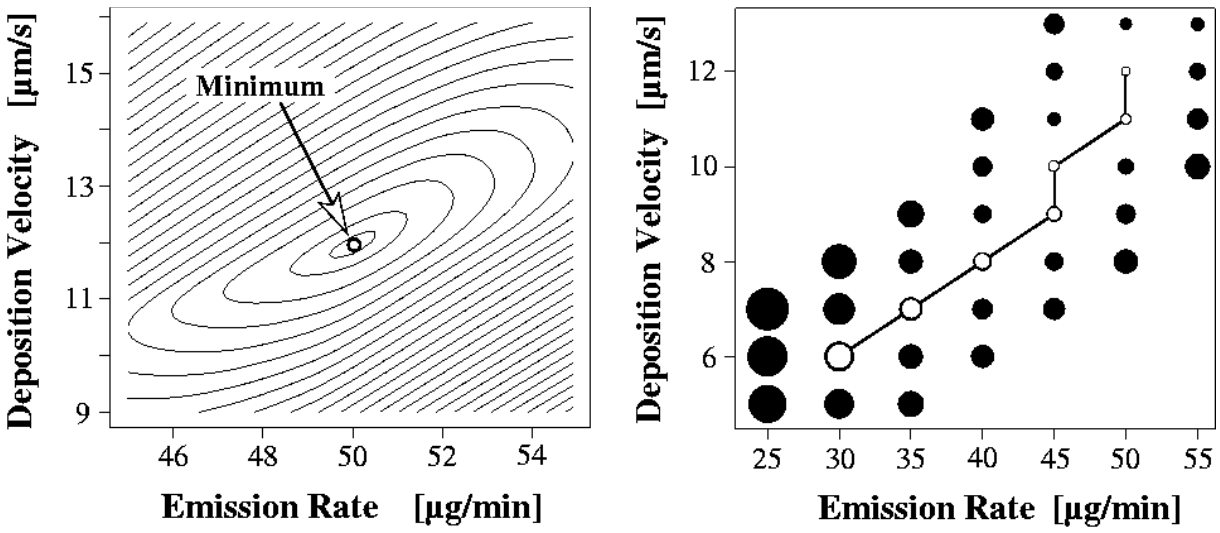

Figure 1. The left plot shows contours of the optimization response surface for particles with diameters of 0.1 - 0.2 µm (for the *Cigarillo #2* experiment) over a range of emission rates and deposition velocities. The right plot depicts the *optimization pathway*, illustrating how the local grid search method was used to find the minimum point on the surface and the optimal values of 12 µm/s and 50 µg/min. The circle size indicates the error of the model in fitting the measurements.

RESULTS AND DISCUSSION

Table 1 contains a summary of each experiment and the filter-based results for TPM emissions. Table 2 contains our best estimates of the size distribution of particle mass emissions and integrated emissions for eight experiments. The total particle mass emitted by the cigarillos and premium cigar and their emission rates were markedly lower than for the other types of cigars and for cigarettes. This finding may be an artifact of leakage around the end-fittings during smoking (the cigarillos had plastic tips and the premium cigar was rather bulky). Total particle emissions determined by integrating the particle mass size distributions (Table 2) were generally lower than those determined using filters (Table 1) with total particle mass emissions ranging from 54 - 84% of the filter-based emissions. The larger values for filters may be a consequence of the collection onto the filters, by sorption or condensation, of semivolatile organic compounds that are present in ETS. The integrated emissions were consistently higher for cigarettes (7 - 8 mg/g-smoked and 0.7 – 0.9 mg/min) than for cigars (3 - 5 mg/g-smoked and 0.2 – 0.7 mg/min). The mass-normalized emissions (mg/g-smoked), which may be more appropriate for direct comparisons, showed consistent results among different types of cigars.

The fits between observed and modeled time series data were generally quite good with minima for the mean absolute deviation in each bin ranging from 0.9 to 3 $\mu g/m^3$. The model seemed to accurately account for the effect of particle coagulation, which influenced concentrations in the smaller size bins for times as long as 4 hours after the source was extinguished. However, we observed small systematic deviations in the fits for the first hour or two of a few of the time series in the > 0.4 µm particle diameter range, where the observed particle loss appeared to be faster than later in the time series. This effect may be due to evaporative losses.

Figure 2 shows the log-normal fits to particle emissions for selected experiments. The fitted parameter values are given in Table 2. MMDs were close to 0.2 μm (mean = 0.20 μm; sd = 0.02 μm) for all source types. The variation in GSDs was larger with a range of 1.9 to 3.1 (mean = 2.3; sd = 0.37). ETS particle mass emissions appear to be due mostly to particles that have diameters between 0.02 and 2 μm — with no clear difference in the estimated mass size distributions for cigars and cigarettes.

Table 1. Summary of chamber experiments and filter-based ETS-particle emissions

Experiment	Smoking Duration [min]	Tobacco Mass Smoked [g]	Filter-Based Emissions[a]		
			TPM [mg]	Rate [mg/min]	Mass-Normalized [mg/g-smoked]
Regular Cigar #1	12.5	1.71	10.8	0.86	6.3
Regular Cigar #2	14.8	1.46	8.1	0.55	5.6
Regular Cigar #3	10.3	1.92	9.5	0.92	4.9
Regular Cigar #4	11.3	1.41	11.8	1.04	8.4
Regular Cigar #5	14.0	1.50	7.3	0.52	4.8
Premium Cigar	13.4	1.26	5.6	0.42	4.0
Cigarillo #1	10.1	1.02	4.0	0.67	6.6
Cigarillo #2	14.8	0.96	4.0	0.27	4.1
Cigarillo #3	14.8	1.36	6.3	0.42	4.6
Cigarette #1	5.5	0.73	9.8	1.79	13.4
Cigarette #2	6.1	0.72	7.0	1.15	9.7
Cigarette #3	7.4	0.72	9.3	1.3	13.0
Cigarette #4	7.1	0.72	7.3	1.03	10.1

[a] Emission rate and mass-normalized emissions were calculated by dividing the total mass emissions by the smoking time and mass of tobacco smoked, respectively.

Table 2. The estimated size distributions of ETS-particle emissions

Experiment	MMD[a] [μm]	GSD[b]	Integrated Emissions[c]		
			TPM [mg]	Rate [mg/min]	Mass-Normalized [mg/g-smoked]
Regular Cigar #1	0.18	2.5	8.8	0.71	5.2
Regular Cigar #2	0.21	2.2	6.7	0.46	4.6
Regular Cigar #3	0.20	2.4	6.3	0.61	3.3
Premium Cigar	0.18	1.9	4.7	0.35	3.7
Cigarillo #2	0.23	3.1	2.8	0.19	2.9
Cigarette #2	0.21	2.1	5.5	0.90	7.7
Cigarette #3	0.20	2.1	5.0	0.68	7.0
Cigarette #4	0.19	2.1	5.1	0.71	7.1

[a]MMD is the fitted mass median diameter, also called the mass geometric mean diameter.
[b]GSD is the fitted geometric standard deviation.
[c]The "integrated'' total mass, rate, and mass-normalized emissions were obtained by integrating the estimated size-distribution of total mass emissions and multiplying by unity, the smoking time, or the mass of tobacco smoked, respectively. See Table 1 for the values of the smoking time and mass of tobacco smoked.

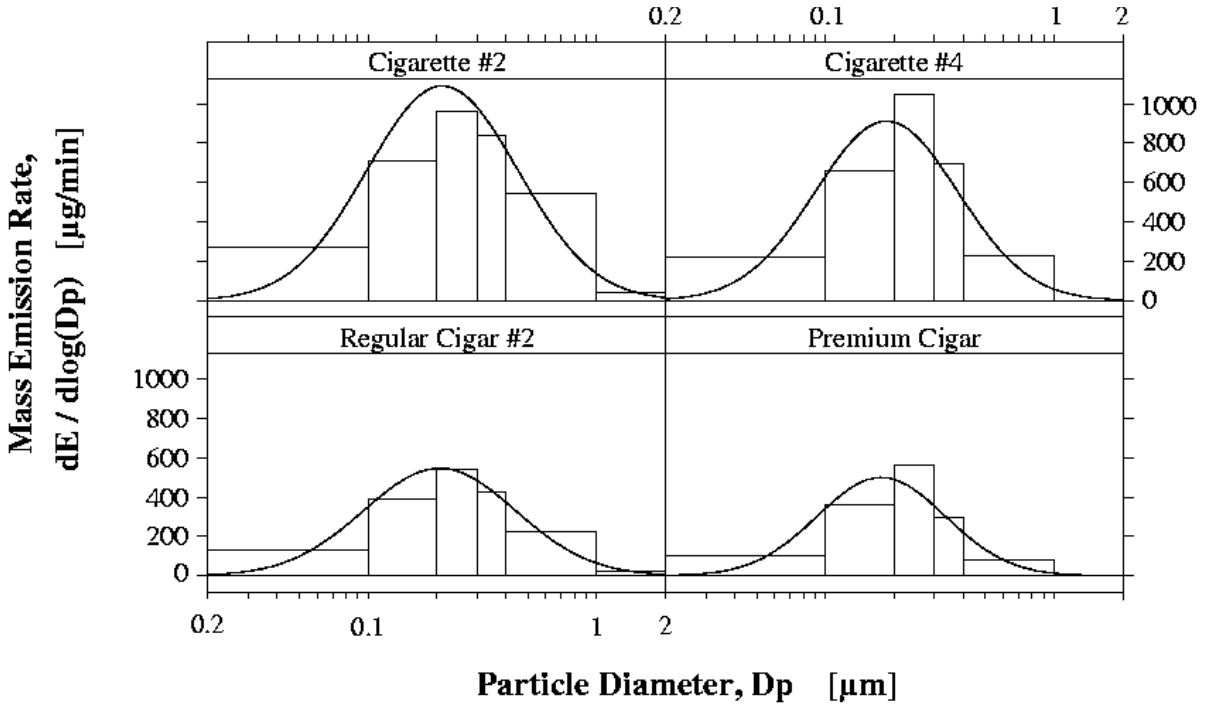

Figure 2. Plots of the estimated size distribution of particle mass emission rate (in µg/min) and the corresponding log-normal fits for four experiments. The text at the top of each panel states the type of source used in each experiment. See Table 2 for the fitted mass median diameter (MMD), geometric standard deviation (GSD), and integrated mass emissions.

Since our approach neglects evaporation, our results may have been influenced by the evaporation of volatile particle constituents, which is likely to have occurred as the emitted ETS was rapidly mixed and diluted with chamber air. The true emissions may be larger in magnitude and occur at larger particle sizes than we determined. However, using the optimal values of emissions and deposition rate for input, our model provides a good fit to ETS-particle concentrations for most size ranges and for most times, and we therefore judge it to be an appropriate tool for predicting concentrations and exposures. In addition, our results for the size distribution of ETS-particle emissions are in generally good agreement with the findings of other investigators. Ingebrethsen and Sears (1989) find an MMD of 0.2 µm for aged and diluted sidestream smoke and Ueno and Peters (1986) find an MMD of 0.16 µm with a GSD of 1.4-1.7. Chang et al. (1985) reported an average MMD of 0.26 µm for diluted mainstream smoke with a GSD of 1.2. In alignment with our findings, both Ueno and Peters and Chang et al. report total mass emissions from real-time instruments (using either electrostatic mobility or light scattering) that are as little as 50% of determinations based on direct mass measurements (a cascade impactor in their case). As far as we know, this discrepancy has not been resolved and will require further investigation.

CONCLUSION AND IMPLICATIONS

ETS particle concentrations depend on the conditions of mixing and dilution, including smoking style, which can vary from one smoking situation to another. Therefore, additional emissions characterization and model evaluation studies, similar to ours, should be carried out using designed experiments that capture real-world variation in room conditions and smoking behavior. In the interest of improving human exposure and risk assessments, the size-

resolved emissions of other indoor particle combustion sources that emit submicron particles, such as heaters, stoves, incense, or candles, should also be studied.

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INHALATION OF HAZARDOUS AIR POLLUTANTS FROM ENVIRONMENTAL TOBACCO SMOKE IN US RESIDENCES

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ABSTRACT
In the United States, 48 million adults smoke 5×10^{11} cigarettes per year. Many cigarettes are smoked in private residences causing regular environmental tobacco smoke (ETS) exposure to at least 31 million nonsmokers (11% of the US population), including 16 million juveniles. ETS contains many chemical species whose industrial emissions are regulated by the US federal government as hazardous air pollutants (HAPs). In this paper, average daily residential exposures to 15 HAPs in ETS are estimated for US nonsmokers who live with smokers. The evaluation is based on material-balance modeling, and utilizes published data on smoking habits, demographics, and housing. Newly measured exposure-relevant emission factors are incorporated. Comparison of exposure concentration estimates with health-based guidelines for chronic exposure suggests that aldehydes — specifically acrolein, acetaldehyde, and formaldehyde — should be of particular concern in ETS. Cumulative population intake results are compared for these compounds against other sources of exposure.

INDEX TERMS
Environmental tobacco smoke, Exposure, Hazardous air pollutants, Health risk, Mass-balance modeling

INTRODUCTION
Cigarette smoking is a serious contributor to indoor air pollution. Exposure to environmental tobacco smoke has been linked to an increased risk of many adverse health outcomes, including lung cancer, asthma onset and exacerbation, and acute respiratory illness (NCI, 1999). Concerns about ETS exposure have led to restrictions on smoking in public places, including an almost complete ban on smoking in enclosed workplaces in California. However, regulatory approaches have limited utility for reducing ETS exposures in private residences. Instead, public education, possibly augmented by technological interventions, is best suited to reduce exposures.

Tobacco smoke comprises a large number of chemical constituents, variously partitioned between the gas and condensed phases. Among the constituents of ETS are chemical compounds that are regulated as hazardous air pollutants (HAPs) by the US federal government (USEPA, 2001a). HAPs are species that are known or suspected carcinogens, or that have been shown to cause other serious health effects, such as reproductive problems or birth defects. Characterization and control of HAPs has focused on outdoor sources (USEPA, 2001b). However, because of the close proximity between smokers and nonsmokers, and because of the persistence of pollutants in indoor spaces, the inhalation intake per unit emission is 100 or more times higher for ETS than for typical outdoor sources (Smith, 1993; Lai et al., 2000).

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To aid in risk assessment, California's Environmental Protection Agency has developed chronic inhalation reference exposure levels (RELs) for 75 individual air toxicants (OEHHA, 2001). A chronic REL represents an estimate of the airborne concentration to which individuals may be indefinitely or routinely exposed with no associated significant health risk. Since ETS is just one of many sources of pollution exposure, chronic exposures associated with ETS near or in excess of the RELs would constitute a cause for concern.

This paper describes an analysis of exposures to specific ETS-constituent HAPs for nonsmokers who live with smokers. Estimated exposure-relevant concentrations are compared to chronic RELs to investigate risks posed by individual HAPs in ETS. The significance of cumulative population exposures is also explored.

OVERVIEW OF APPROACH

The overall approach adopted here combines a material balance model with published and newly generated data for key input variables. The material balance model is used to estimate exposure concentrations based on data or estimates for these parameters: cigarette consumption patterns, emission factors for HAPs from ETS, residence volume and air-exchange rates, and population statistics. The exposed populations of interest here are nonsmoking adults and all juveniles who live with smokers. A primary goal is to estimate the central tendency of daily exposure to specific hazardous air pollutants by members of this exposed population. The cumulative intake by the entire population will also be estimated. Health risk information will be considered to identify the specific contaminants, among those assessed, which pose the greatest health risk. The health concerns considered here are long-term risks associated with chronic exposure, such as cancer, rather than acute concerns such as odor and irritation.

CIGARETTE SMOKING HABITS IN THE UNITED STATES

The prevalence of smoking among noninstitutionalized US adults was determined from the Behavioral Risk Factor Surveillance System (BRFSS). This is a state-based, random-digit-dialed telephone survey. Current smokers are defined as those who reported having smoked more than 100 cigarettes during their lifetime and who currently smoke every day or on some days. We combined state-by-state, gender-specific information from BRFSS with census data to estimate that 47.7 million adults (24.9 million males) currently smoke cigarettes in the US (MMWR, 2001; US Census Bureau, 2002), an overall adult smoking prevalence of 22.8%.

The quantity of cigarettes consumed by smokers was estimated from 1997 records reporting an adult, per capita, tax-paid sales rate of 117 packs per year (Tobacco Institute, 1997). Assuming this rate applies to the current US adult population of 213 million, we estimate an annual consumption of about half a trillion (4.9×10^{11}) cigarettes by US adult smokers, which corresponds to 1.4 packs per day per smoker.

HOW MANY PEOPLE ARE EXPOSED TO ETS AT HOME?

We next seek to estimate the number of nonsmokers who are regularly exposed to environmental tobacco smoke (ETS) in their residences. Comprehensive, unbiased estimators do not exist; however, an estimate can be constructed from good proxies. The Third National Health and Nutrition Examination Survey (NHANES III) collected data during 1988-1991 on a nationally representative cross-section of 7079 juveniles (aged 2 mo to 16 y) and 9769 adults (aged 17 and older). In this study, exposure to ETS at home was assumed to occur if any household member smoked, a condition that was reported for 40.8% of nonsmoking juveniles

and 17.4% of nonsmoking adults (Pirkle et al., 1996). This survey likely overestimates the prevalence of household ETS exposure, since it does not exclude homes with smokers who do not smoke indoors. As part of the BRFSS in 1996, data were collected on the prevalence of households with current adult cigarette smokers and any children and adolescents in the home (MMWR, 1997). The same investigation collected information on whether smoking was permitted in some or all areas of the home. From this information, the number of juveniles exposed to ETS at home was estimated to be 15.7 million, or about 22% of all US juveniles. Assuming that the same proportion (22%/40.8% = 0.54) can be applied to the NHANES III adult data, then the proportion of nonsmoking adults regularly exposed to ETS in their own homes is estimated to be 9.4%, which corresponds to 15.6 million people. Thus, we estimate that about 31 million nonsmokers are regularly exposed to ETS in their own homes because they live with smokers, and half of those exposed are children and adolescents.

EXPOSURE-RELEVANT EMISSION FACTORS

Important input into this analysis is the effective rate at which each of the HAPs is emitted in ETS when a cigarette is smoked. Emission factors have been measured for many ETS constituents in special test chambers. However, recent work indicates that airborne concentrations of some ETS constituents can be greatly affected by sorptive interactions with indoor surfaces (Singer et al., 2002a). Sorption can reduce concentrations and short-term exposures relative to those predicted using an emission factor measured under conditions of low sorption (e.g., an unfurnished metal chamber). We address this issue with the concept of an exposure-relevant emission factor (EREF) that implicitly incorporates sorption effects under realistic furnishing and ventilation conditions. For this analysis, we are interested in ETS concentrations that result from conditions of regular daily smoking. Making use of data from a recent experimental investigation of ETS gas-phase dynamics (Singer et al., 2002b), we derived EREFs by mass balance from gas-phase ETS concentrations measured during the fourth week of an experiment in which 10 cigarettes were smoked each day, 6 days per week, in a furnished 50-m^3 room ventilated at 0.6 h^{-1}. Calculated EREFs are reported in Table 1.

QUANTIFYING EXPOSURES TO INDIVIDUAL ETS CONSTITUENTS

In keeping with the goal of providing an assessment that is objective, transparent, and as accurate as the empirical data will support, the daily residential exposure to ETS by nonsmokers is estimated by means of a simple material-balance model. We define the following variables: N is the mean number of cigarettes smoked per day in the home (cig d^{-1}); V is the building volume (m^3); A is the number of indoor-outdoor air exchanges per day (d^{-1}); and E_i is an exposure-relevant emission factor for the HAP of concern (µg cig^{-1}). The daily average concentration of the HAP caused by ETS for homes where smoking occurs is estimated as

$$C_i = \frac{NE_i}{AV} \qquad (1)$$

To generate central-tendency estimates for C_i, parameter values that approximate the expected mean are selected for terms on the right-hand side of equation (1). We take N = 20 cig d^{-1} (= 1 pack per day) on the basis that people in the US spend an average of 69% of their time indoors at home (Klepeis et al., 2001), half of which is assumed to be awake; the average number of smokers in a home that includes a nonsmoker is 1.4 (Pirkle et al., 1996); and the average cigarette consumption rate is 1.4 packs per smoker per day. The daily number of indoor-outdoor air exchanges is taken to be A = 15 d^{-1}, based on a nationwide compilation of

air-exchange rate data (Murray and Burmaster, 1995). The residence volume is taken to be 400 m^3, which corresponds approximately to the median floor area (1730 ft^2) reported for US single-family detached and mobile homes (US Census Bureau, 2001).

To facilitate our analysis, we assume that the average concentration of an ETS constituent in air breathed by the nonsmoker over the course of a day, caused by smoking in their home, is the same as the average in ventilation air that passes through the residence. This is approximately the condition that would obtain if the nonsmoker were present in the home during, and for several hours after cigarette consumption by the smoking resident(s). To the extent that nonsmokers are not present during and after smoking, the exposure would be reduced. Conversely, if the nonsmoker were present in the same room as the smoker during smoking events, the exposure concentration would probably be higher than estimated here. Table 1 presents an estimate of daily average exposure concentrations for 15 HAPs and compares these results with chronic RELs, where those are available. The rightmost column of Table 1 shows the ratio of estimated exposure concentration to the chronic REL. The highest value of this ratio, ~ 30, is found for acrolein. Values in the vicinity of 1 are found for the other aldehydes: acetaldehyde and formaldehyde. That these ratios are of order one or greater than one, and that these are the highest ratios, suggests that, among the compounds considered here, exposure to aldehydes from ETS should be of particular concern.

Table 1. Central-tendency estimates of daily-average exposure concentration to HAPs in ETS for nonsmokers who live with a smoker.

Compound	EREF ($\mu g\ cig^{-1}$) [a]	Exposure conc. ($\mu g\ m^{-3}$)	Chronic REL ($\mu g\ m^{-3}$) [b]	Exposure/REL
Acetaldehyde	**2267**	**7.6**	**9**	**0.8**
Acetonitrile	1028	3.4	na	—
Acrolein	**560**	**1.9**	**0.06**	**31**
Acrylonitrile	181	0.6	5	0.1
Benzene	417	1.4	60	0.02
1,3-Butadiene	461	1.5	20	0.08
2-Butanone	300	1.0	na	—
Cresol isomers	70	0.2	600	0.0003
Ethylbenzene	136	0.5	2000	0.0002
Formaldehyde	**979**	**3.3**	**3**	**1.1**
Methylnaphthalenes [c]	52	0.2	na	—
Naphthalene	42	0.1	9	0.02
Phenol	162	0.5	200	0.003
Styrene	169	0.6	900	0.0006
Toluene	879	2.9	300	0.01

[a] Exposure-relevant emission factor; see text for details.
[b] Reference exposure level for chronic conditions (OEHHA, 2001).
[c] Sum of 1-methylnaphthalene and 2-methylnaphthalene isomers.

INHALATION INTAKE OF ALDEHYDES FROM ETS

In this section, the total annual inhalation intake of ETS-associated aldehydes by US nonsmokers who live with smokers is estimated. To put the result in context, an estimate is

also made of the total inhalation intake by all US residents from all primary outdoor emissions.

For the 31 million nonsmokers who live with smokers, the estimated ETS-associated exposure concentration of acrolein is 1.9 $\mu g\ m^{-3}$. The average volume of air breathed per day is 12 m^3 (Layton, 1993). Multiplying these three numbers together and then converting to mass inhaled per year, we estimate the total intake of acrolein by this population to be 260 $kg\ y^{-1}$. This corresponds to about 0.1% of the total of 280 tonnes of acrolein generated in ETS each year.

The total primary emissions of acrolein to the atmosphere from all sources in the United States are about 26,000 tonnes per year, according to the Toxic Release Inventory (USEPA, 2001b). Most of this is from area sources that would tend to occur in populated regions. The fraction of pollution emitted to urban air that is breathed by people has been estimated to lie in the range 0.7-70 per million, depending on meteorology and on the size of the populated region, and assuming a typical urban population density of 1000 km^{-2} (Lai et al., 2000). Taking 10 per million as a crude estimate of the average condition in the United States, we estimate that the total inhalation intake of acrolein from all ambient emission sources is ~ 300 $kg\ y^{-1}$. Thus, although ETS is almost negligible as a source of urban air concentrations of acrolein, it contributes as much to cumulative human intake in residential settings alone as do all sources of emissions to the ambient air. For acetaldehyde, a similar conclusion is reached: ETS in homes contributes about as much to human inhalation intake (~ 1000 kg/y) as do all primary-emission sources to ambient air. On the other hand, formaldehyde emissions to ambient air are estimated to be stronger contributors to human inhalation exposure (~ 3000 kg/y) than ETS in homes (~ 400 kg/y).

CONCLUSIONS

Most health-risk evaluations of exposure to environmental tobacco smoke are based on epidemiological investigations that use questionnaires or marker compounds (e.g., cotinine in body fluids) to estimate exposure. The approach presented here is complementary: it can help to identify specific compounds in ETS that contribute significantly to the overall health risks. The results of our study indicate that the aldehydes in ETS, especially acrolein, should be of particular concern as contributors to health risk from chronic, residential ETS exposure. Future studies may usefully be focused on better characterizing exposure to aldehydes from ETS and on the effectiveness of intervention measures to reduce such exposures.

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EFFECT OF SORPTION ON EXPOSURES TO ORGANIC GASES FROM ENVIRONMENTAL TOBACCO SMOKE (ETS)

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ABSTRACT
The effects of sorption processes on dynamic ETS organic gas concentrations and potential exposures were studied in a carpeted and furnished 50-m^3 room ventilated at 0.6 h^{-1}. Ten cigarettes were machine-smoked on six of every seven days over four weeks. Concentrations of ETS-specific tracers and regulated toxic compounds were quantified during daily smoking, post-smoking and background periods. Potential exposures were calculated by period and day. Large sorption effects were observed for the widely used tracers 3-ethenylpyridine and nicotine, and for several toxic compounds including naphthalene and cresol isomers. Short-term adsorption to indoor surfaces reduced concentrations and potential exposures during smoking, while later reemission increased concentrations and exposures hours after smoking ended. Concentrations during nonsmoking periods rose from day to day over the first few weeks, presumably from increased reemission associated with increased sorbed mass concentrations. For sorbing compounds, more than half of daily potential exposures occurred during nonsmoking periods.

INDEX TERMS
Environmental tobacco smoke, Pollutant sorption and desorption, Exposure assessment, VOCs and SVOCs, Laboratory and field experiments

INTRODUCTION
Environmental tobacco smoke (ETS) is a dynamic mixture of particles, inorganic compounds, and organic gases that span a wide volatility range. The ETS gas-phase (filtered ETS) has been identified as carcinogenic in animal-based toxicology studies (Witschi et al., 1997). Many individual ETS organic gases are regulated as toxic air contaminants and hazardous air pollutants (henceforth referred to as “toxic compounds”) by the California state and United States federal governments, respectively (CARB, 2001).

Quantifying exposures to toxic organic gases in ETS is challenging since many of these compounds are also emitted from other sources. One approach is to measure exposure to a tracer compound that is specific to ETS. Exposure to ETS toxic components then may be estimated based on the relative abundances of toxic and tracer compounds as measured in ETS under controlled conditions. This approach requires that tracer and toxic compounds exhibit similar dynamic behavior (Daisey, 1999). Nicotine and its primary metabolite cotinine are the most widely used chemical tracers of ETS exposure. Yet it has been known for some time that nicotine dynamics differ from those of other ETS components (Eatough, 1993). In light of this concern, other gas-phase tracers have been suggested, including 3-ethenylpyridine (3-EP), pyridine, and pyrrole (Eatough, 1993; Hodgson et al., 1996).

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Concentrations of nicotine, 3-EP, and some toxic ETS compounds are affected by sorption (Piade et al., 1999; Van Loy et al., 2001). Adsorption to indoor surfaces can reduce gas-phase concentrations during smoking events, while later reemission (desorption) can raise concentrations long after smoking has stopped. If sorption is reversible and reemission occurs on a time scale of hours or days, the sole effect should be to shift potential exposures in time. By contrast, if some fraction of the mass is sorbed irreversibly, or the time scale of reemission is much slower, then sorption should reduce potential short-term exposures associated with a single emission event. We hypothesize that when smoking occurs regularly, e.g. on a daily basis, sorbing organic compounds will accumulate on surfaces. Higher surface concentrations of reversibly sorbed mass should, in turn, lead to increased reemission rates and higher gas-phase concentrations during nonsmoking periods. Thus, we expect that longer-term sorption processes will increase potential exposures beyond those resulting from new emission events.

We investigated the dynamic behavior of ETS organic gases through experiments in which cigarettes were smoked by machine in a facility designed to simulate residential and office environments. Key components that affect sorption, such as ventilation rate and furnishing level (i.e. material type and surface area) were varied. A detailed investigation of sorption effects on exposures for single-day ETS emission events is reported elsewhere (Singer et al., 2002). This paper presents preliminary results from an investigation of sorption effects on exposures when smoking occurs on a repeated daily basis.

METHODS

Experiments were conducted in a 50-m^3 room that was constructed and furnished with typical indoor materials including painted gypsum wallboard; nylon carpet; upholstered chairs; wood and veneer desks, bookcases and tables; and heavy cotton draperies. The floor (carpet) area was 20 m^2 and the projected area of the chairs, draperies and wood furniture was 43 m^2. With 84 m^2 of wallboard, the total surface-to-volume ratio was 2.6 m^{-1}. ETS was simulated with the sidestream emissions from machine smoking of a popular brand of filter cigarettes. Room ventilation air passed through a bed of activated carbon to reduce organic gas concentrations in the inlet air. Air samples were collected on sorbent tubes containing Tenax-TA and Carbosieve S-III 60/80 mesh. Organic compounds were analyzed by thermal desorption gas chromatography/mass spectrometry. Target compounds included regulated toxics, ETS tracers, and other prominent organic gases in ETS. Detailed descriptions of all methods are provided elsewhere (Singer et al., 2002).

The effects of short- and long-term sorption processes on dynamic ETS organic gas concentrations and potential exposures were studied through two types of experiments. In the first set, organic gas concentrations were measured over multi-hour periods for 3-4 days following a single 3-h smoking period. The primary goal was to quantify the effect of sorption processes on exposures to new emissions of ETS. Exposure-relevant emission factors that account for sorptive losses and re-emission over a 24-h period were determined for three furnishing levels (wallboard with aluminum flooring, wallboard with carpet, wallboard with carpet and full furnishings) and three ventilation rates (nominally 0.3, 0.6 and 2.0 h^{-1}). Full results have been reported (Singer et al., 2002). An ongoing second group of experiments focuses on the effect of longer-term sorption and reemission processes that pertain when smoking occurs regularly and repeatedly. This paper presents results from an experiment in which 10 cigarettes were smoked in the fully furnished room during a 3-h period each day, six days per week, for 4 weeks, with a chamber ventilation rate of 0.6 h^{-1}. Organic gas concentrations were measured over three daily periods: a 4 h “smoking” period extending for

1 h after smoking ended, a 10 h "post-smoking" period, and a 10 h daily "background" period. These periods were chosen to simulate a residential scenario in which a working adult smokes for several hours each evening. Samples were collected during the background period each day, and during each period on days 1, 3, 6, 10, 13, 17, 20, 24 and 27.

RESULTS AND DISCUSSION

The short-term effects of sorption can be seen in Figure 1, which shows potential daily exposures to seven representative ETS gases on Day 1 of the repeated daily smoking experiment. These are potential exposures that would occur for a nonsmoker occupying the room throughout each period. Potential exposures were calculated as the product of the measured gas-phase concentrations and the duration of the period. For 1,3-butadiene and benzene, time-dependent concentrations were controlled entirely by emissions and ventilation; about 73% of the potential exposure occurred during the 4-h smoking period with the remaining 27% during the next 10 h period. By contrast, potential exposures during the smoking period comprised only about half of the daily total for the regulated toxics p-cresol (48%) and naphthalene (55%), and for the ETS tracer 3-EP (48%). Potential exposures during the post-smoking period accounted for 30-38% of daily totals for these compounds. The remaining 14-18% was attributed to the background period. Short-term sorption and reemission processes thus shifted about one quarter of potential exposures of these three compounds from smoking to nonsmoking periods. The ETS tracer pyridine and the toxic compound styrene (not shown) exhibited intermediate behavior with potential exposures distributed by period as follows: smoking (65%), post-smoking (35%), and background (5%). For pyrrole (also not shown), potential exposures were split evenly between smoking and post-smoking periods (45% each). The small fraction of nicotine exposure during nonsmoking periods indicates a slower, or less-reversible sorption.

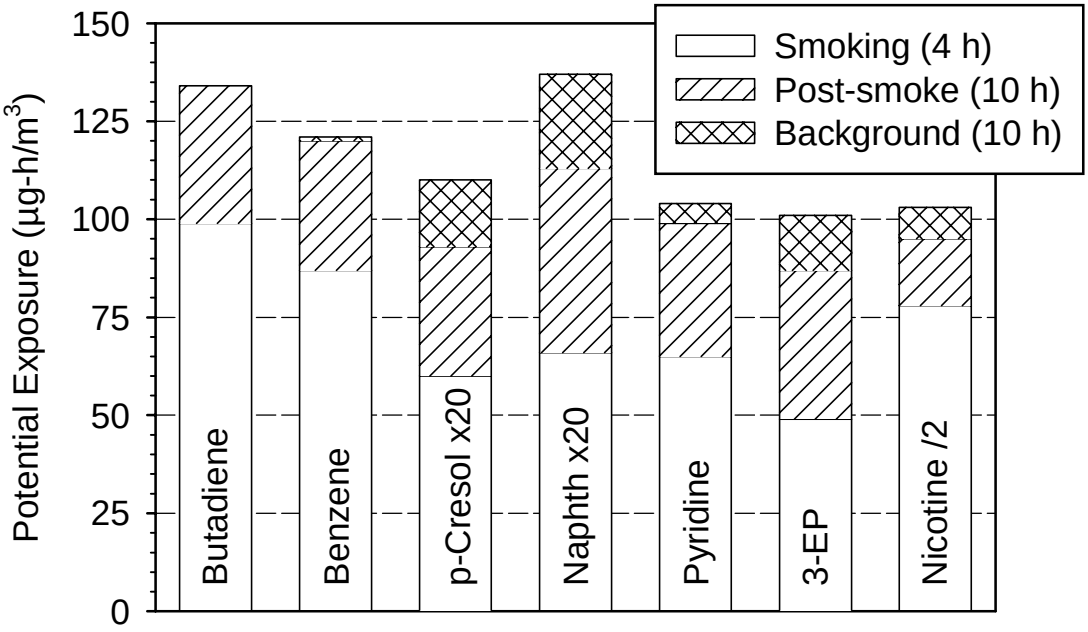

Figure 1. Potential ETS exposure, by period of day, in a furnished 50-m^3 room on **Day 1** of the repeated daily smoking experiment. 10 cigarettes were smoked by machine during the first 3 h of the designated smoking period. Naphth = Naphthalene; 3-EP = 3-Ethenylpyridine.

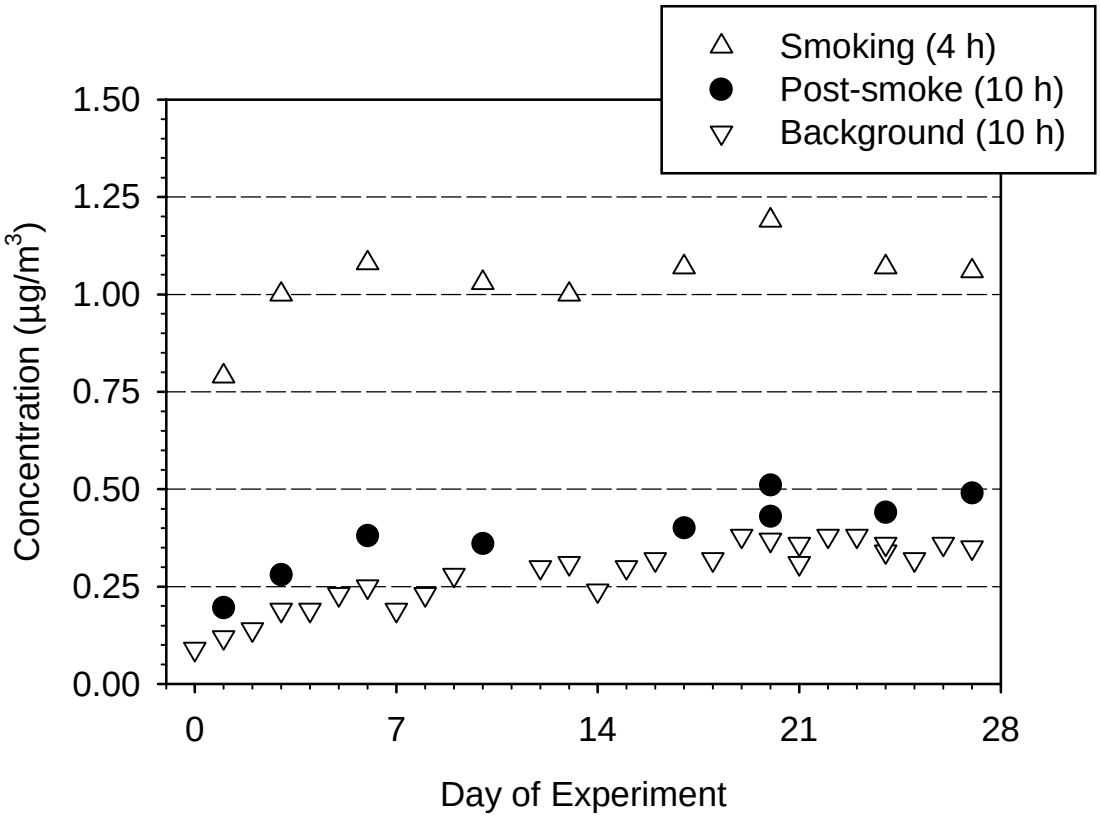

Figure 2. Time-averaged concentrations of p-cresol measured during smoking, post-smoking and background periods in a 50-m^3 furnished chamber. 10 cigarettes were smoked each day, six days per week over a 4-week period. No cigarettes were smoked on days 7, 14, and 21.

Mass loss due to longer-term sorption necessarily leads to the accumulation of sorbed mass on indoor surfaces. To the extent that the sorption is reversible there should be a consequent increase in reemission; this should in turn lead to higher gas-phase concentrations of the sorbing compounds over time. Figure 2 demonstrates this phenomenon for p-cresol over the 4-week repeated daily smoking experiment. Background period concentrations generally rose on each smoking day of the first two weeks, but dropped on days without new ETS emissions. Background concentrations leveled off during the third week then fluctuated from day to day, but did not increase overall during the fourth week. The rise in concentrations during daily smoking and post-smoking periods mirrored the rise during background periods.

These higher concentrations translate into higher potential ETS exposures for sorbing compounds when smoking is habitual. Figure 3 shows potential exposures on Day 27, resolved by daily period, for the same compounds shown in Figure 1. A comparison of the total daily exposures by compound indicates that for 1,3-butadiene and benzene, potential exposures were essentially unaffected by long-term sorption and reemission processes. (The increases of 8% for 1,3-butadiene and 20% for benzene may result from emission variability.) By contrast, potential daily exposure increased by about 110% for naphthalene and p-cresol, by 80% for 3-EP and by 180% for nicotine. Results for phenol and other cresol isomers were similar to those shown for p-cresol, whereas larger increases of 150% were observed for methylnaphthalene isomers. This result suggests that even larger effects are likely for certain polycyclic aromatic hydrocarbons (PAH), primarily three-ring compounds, that are larger than the methylnaphthalenes but still volatile enough to exist primarily in the gas-phase of

ETS. Reemission of long-sorbed mass increased potential exposures to other ETS tracers by only 30% for pyridine and by 35% for pyrrole (not shown in figure).

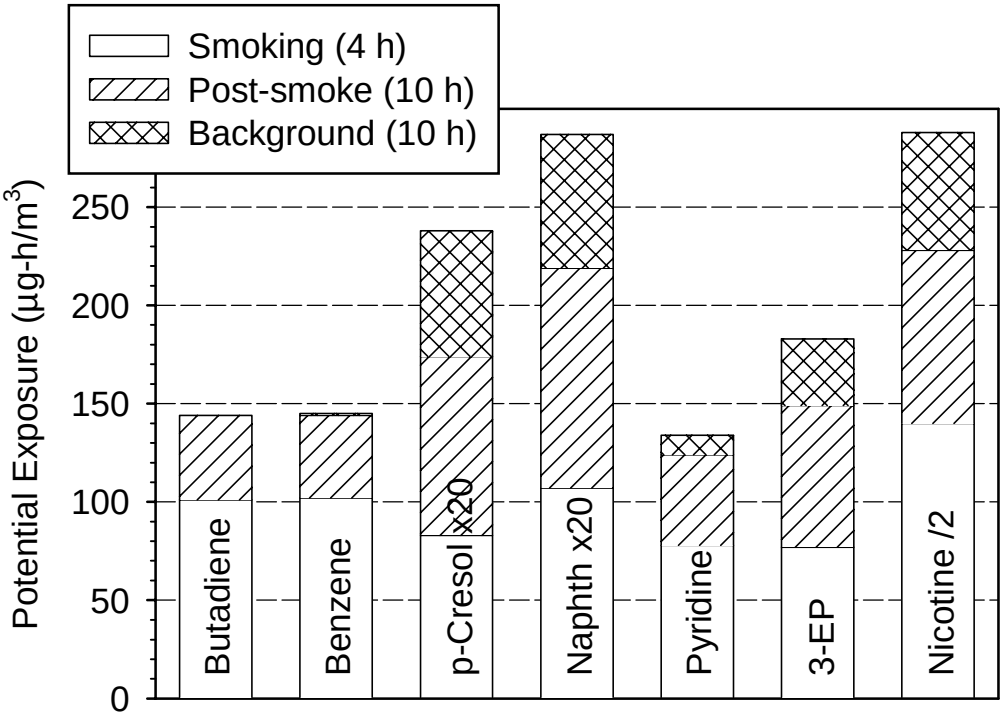

Figure 3. Potential ETS exposure, by period of day, in a furnished 50-m^3 room on **Day 27** of the repeated daily smoking experiment. 10 cigarettes were smoked by machine during the first 3 h of the designated smoking period. Naphth = Naphthalene; 3-EP = 3-Ethenylpyridine.

Figure 3 also shows that increased reemission rates shifted a larger fraction of potential daily exposures from smoking to nonsmoking periods. Potential exposures for 1,3-butadiene and benzene again occurred mostly (70%) during smoking periods, with the remaining 30% during the nominal post-smoking period. For the sorbing toxic compounds p-cresol and naphthalene, the smoking period accounted for only 35% of potential daily exposures; 40% occurred during the post-smoking period and 25% was attributed to the background period. Similar results were obtained for 3-EP and for pyrrole (not shown). The fraction of potential daily exposure during the smoking period was again higher for nicotine compared to the other sorbing compounds. But by the end of the fourth week, only about half of the potential daily nicotine exposure could be attributed to the smoking period, with 30% and 20% occurring during the post-smoking and background periods, respectively. Despite the overall increase in daily exposure potential for pyridine and pyrrole, the distribution of potential exposures throughout the day did not change appreciably for these compounds over the 4-week experiment. These results suggest that among the compounds studied, pyridine may be the best tracer of the dynamics of non-sorbing ETS compounds.

CONCLUSION AND IMPLICATIONS

The experimental data presented in this paper substantiate the hypothesized effects described in the introduction, and provide an initial estimate of the magnitude of these effects in typical residences. Overall exposures will be lower in most circumstances, since the volume of our experimental room is only about 10-20% of a typical home volume and our smoking rate was higher than would be expected for most smokers. But the observed linearity of sorption effects across varying ETS concentration levels (Singer et al., 2002) suggests that the effects

reported here should apply to the lower concentrations that prevail when ETS is diluted throughout a residence. Surface-to-volume ratios also vary, but the quantity and types of surfaces in our test room are typical enough that our results should be generally indicative of sorption effects in real residential environments. Longer-term sorption effects at lower ETS concentrations are being examined using data from an experiment in which 5 cigarettes were smoked daily for 4 weeks with the room ventilated at 2.0^{-1}.

These experimental results provide the most complete picture to date of the range of effects that sorption processes may have on indoor exposures to organic gases in general and to ETS compounds in particular. They indicate that exposures to ETS organic gases under daily smoking conditions can differ markedly from what might be predicted using emission factors or toxic-to-tracer ratios measured in specialized test chambers. A logical next step is to validate these results in a real residence where human smoking or another regular source of organic gas emissions is present. Assessing the implications of these findings to ETS exposures in real-life scenarios will require incorporating information about two additional variables: human activity patterns and inter-zonal transport. Continued efforts along these lines promise to improve our understanding of exposure to constituents of ETS, and also to other sources of volatile and semi-volatile organic compounds.

ACKNOWLEDGEMENTS

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EFFECT OF GASEOUS AMMONIA ON NICOTINE SORPTION

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ABSTRACT
Nicotine is a major constituent of environmental tobacco smoke. Sorptive interactions of nicotine with indoor surfaces can substantially alter indoor concentrations. The phenomenon is poorly understood, including whether sorption is fully reversible or partially irreversible. We hypothesize that acid-base chemistry on indoor surfaces might contribute to the apparent irreversibility of nicotine sorption under some circumstances. Specifically, we suggest that nicotine may become protonated on surfaces, markedly reducing its vapor pressure. If so, subsequent exposure of the surface to gaseous ammonia, a common base, could raise the surface pH, causing deprotonation and desorption of nicotine from surfaces. A series of experiments was conducted to explore the effect of ammonia on nicotine sorption to and reemission from surfaces. Our results indicate that, under some conditions, exposure to gaseous ammonia can substantially increase the rate of desorption of previously sorbed nicotine from common indoor surface materials.

KEY WORDS
Pollutant sorption and desorption, Indoor air chemistry, Environmental tobacco smoke, Laboratory and field experiments, VOCs and SVOCs

INTRODUCTION
Nicotine is a major organic constituent of environmental tobacco smoke (ETS). Typical time-averaged concentrations of nicotine in homes with smokers are ~ 2-4 $\mu g/m^3$ (Coultas et al., 1990; Leaderer and Hammond, 1991). Nicotine is also a strongly sorbing compound. Experiments have shown that sorption onto indoor surfaces significantly reduces concentrations that result from recent emissions (Singer et al., 2002). Gradual reemission of sorbed nicotine could increase exposures during periods without smoking.

Recent studies of nicotine interactions with surface materials have explored the sorption process (Piade et al., 1999; Van Loy et al., 2001); however, it is unclear from the existing evidence whether uptake is fully reversible or partially irreversible. For assessing exposure to nicotine, and for using it as a tracer of exposure to environmental tobacco smoke, the implications of partially irreversible versus fully reversible sorption are large.

Nicotine is a diprotic base that is emitted in the particle phase of sidestream smoke, but quickly partitions to the gas-phase in environmental tobacco smoke (Neurath et al., 1991). Pankow et al. (1997) explored the significance of acid-base chemistry on the portioning of nicotine between the gas and particle phases in mainstream cigarette smoke. They suggested that ammonia plays a key role in influencing nicotine phase partitioning. Nicotine, which initially is bound to the particle phase in a lower volatility, monoprotonated form is thought to lose the proton as pH rises due to the effect of ammonia. The higher volatility, nonprotonated nicotine then partitions more readily to the gas phase.

This line of reasoning led us to consider whether a similar process might explain the process by which nicotine associated with environmental tobacco smoke sorbs to indoor surfaces. We hypothesize that nonprotonated nicotine may sorb to surfaces when ETS concentrations are high and then become protonated in an acid-base surface reaction. The less volatile form bound to the surface would appear to be irreversibly sorbed; however, the process may be reversed by reactions with ammonia (or other bases) that increase surface pH. Nonprotonated nicotine could then desorb more readily from the surface and re-enter the gas-phase.

Ammonia is the most common basic gas in the atmosphere, and major indoor sources include human activities, household cleaners, and litter boxes. Indoor concentrations in homes are typically in the ppb range but can reach tens of ppm during the use of ammonia-based cleaners (Atkins and Lee, 1993).

This paper presents our initial investigation into the effect of ammonia on sorptive interactions between nicotine and indoor surfaces.

EXPERIMENTAL METHODS

Two sets of experiments were conducted in a small glass chamber with a volume of 60 L and a surface area of approximately 1 m^2. In the first experiment, all of the surfaces were lined with a layer of gypsum wallboard that had been coated with low-VOC latex paint. In the second experiment, the wallboard was removed and all surfaces of the chamber were lined with nylon carpet (without any cushion).

Chamber air temperature was controlled between 20-24 °C and relative humidity was in the range of 35-55% during all experiments. Clean dry air was supplied to the chamber from a gas cylinder, and the flow was split such that part of the flow passed through a humidifier to obtain the desired range of relative humidity. The air exchange rate was approximately 0.5 h^{-1} during all experiments.

After pre-conditioning the chamber with clean air and measuring background concentrations, nicotine was introduced using a diffusion tube held at a constant temperature of 30 °C. The diffusion rate was approximately 50 ng/min in a 50 ml/min flow of nitrogen. Nicotine was supplied until concentrations inside the chamber stabilized, indicating that a dynamic equilibrium between sorption and desorption had been reached. The nicotine source was removed, and nicotine concentrations were monitored until they stabilized again. Ammonia was then introduced into the chamber using a permeation tube held at a constant temperature of 30 °C. The supply rate was approximately 2500 ng/min in a 100 ml/min flow of nitrogen. Both the wallboard and carpet experiments were conducted in this manner except that in the carpet experiment the nicotine source was not removed before introducing ammonia.

During the experiments, nicotine and ammonia samples were collected on sorbent tubes inserted through ports in the chamber wall. Air was sampled at a rate of 100 ml/min using pumps and mass-flow controllers located outside of the chamber. Nicotine samples were collected on Tenax sorbent tubes that were thermally desorbed and analyzed by GC/MS. Ammonia samples were collected on sulfuric-acid-treated, silica gel samplers that were extracted and subsequently analyzed using an ammonia-sensing electrode. When duplicate samples were obtained, the results were averaged to calculate concentrations.

A set of three additional experiments was conducted in a room-size chamber furnished with various residential materials. The chamber had a volume of 49.5 m^3, floor dimensions of 4.61 m by 4.42 m, and a height of 2.43 m. During the first two experiments, the chamber was fully furnished (FF). The ceiling and walls were lined with 64.2 m^2 of gypsum wallboard finished with low-VOC latex paint, and the floor was lined with 20.4 m^2 of residential nylon carpet. The furnishings had a total surface area of approximately 43.2 m^2 and included draperies, wood and wood-laminate pieces, and upholstered chairs. During the third experiment, all furnishings were removed leaving only the wallboard and carpet (WBC).

Prior to the experiments reported here, the room-sized chamber had been exposed to ETS. The surfaces were thereby preconditioned with approximately 3200 mg of nicotine sorbed to all surfaces. Background samples of nicotine and ammonia were collected before beginning each set of experiments. All samples were collected and analyzed in the manner described earlier. Chamber air temperature and relative humidity were not controlled and were in the range of 20-24 °C and 33-56%, respectively, during experiments. Clean air was supplied to the chamber by drawing outside air through activated carbon, and the ventilation rate was held constant at 0.3 h^{-1}. Four small fans were present in the chamber to mix the air.

After measuring background concentrations of nicotine, ammonia was introduced into the chamber by diluting 0.24 L (2 cups) of a household ammonia cleaning solution in 3.8 L (1 gal.) of hot water. The bucket containing the mixture was placed in the center of the chamber, and nicotine and ammonia samples were collected for the following three days. At the end of the third day, the bucket was removed and the chamber was flushed with clean air for four days or more before beginning the next set of experiments.

RESULTS AND DISCUSSION

Results from experiments conducted in the small chamber are presented in Figures 1 and 2.

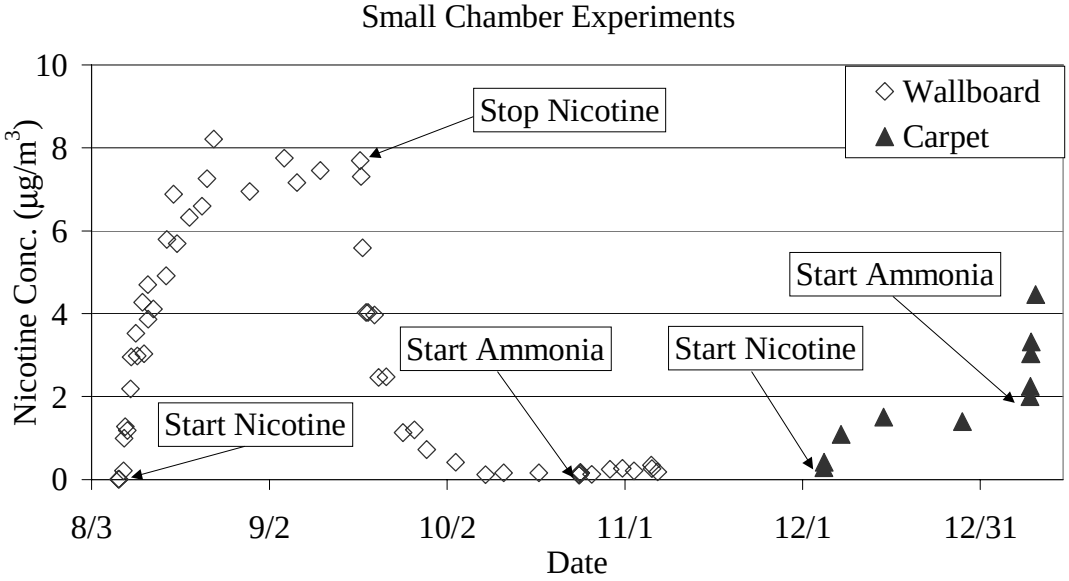

Figure 1. Nicotine concentrations in small chamber during wallboard and carpet experiments

In the first experiment, the chamber was lined with painted gypsum wallboard and the nicotine concentrations stabilized at ~ 8 µg/m^3 six weeks after first introducing the nicotine into the chamber. During this time, the nicotine was sorbing to the wallboard surfaces thereby

reducing the observed airborne concentrations from ~ 100 μg/m^3 that would be expected if sorption were not occurring. After six weeks, the nicotine source was removed and concentrations inside the chamber declined to ~ 0.1 μg/m^3 over the course of another four weeks. Ammonia was then introduced to the chamber. In this case, there was no noticeable increase in nicotine concentrations. Ammonia concentrations remained below 1.5 ppm, indicating a net loss to surfaces, since the level in supply air was ~ 7 ppm. This result suggests that the wallboard material provided a sink for both the nicotine and ammonia and that in this case nicotine desorption was unaffected by acid-base chemistry.

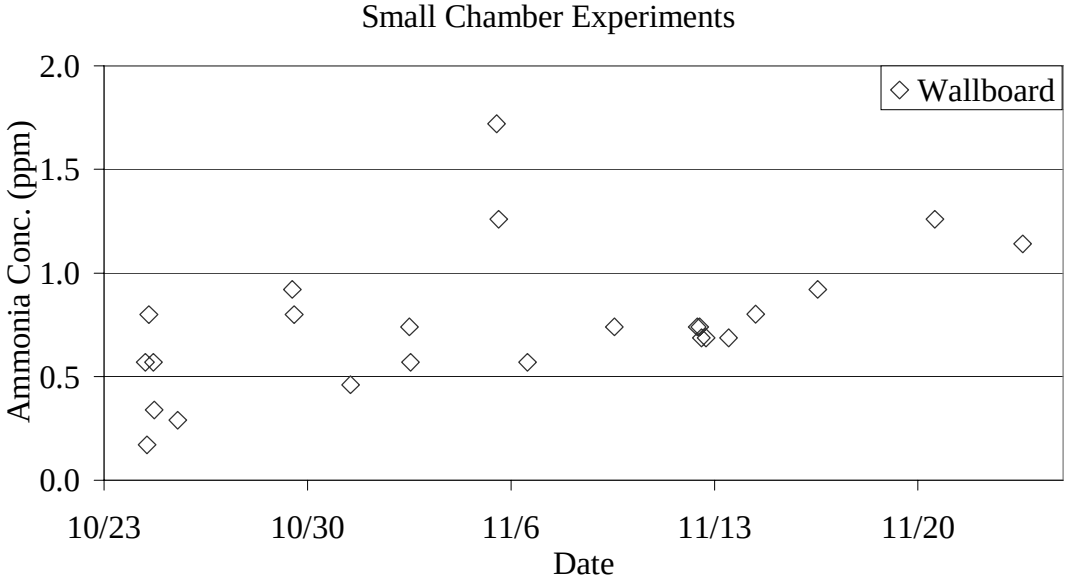

Figure 2. Ammonia concentrations in small chamber during wallboard experiment

In the second experiment, the chamber was lined with nylon carpet. Once again, sorption of nicotine to the surface material reduced airborne concentrations from the expected value of 100 μg/m^3 without sorption to ~ 2 μg/m^3, even after four weeks. This time the nicotine source was not removed before the ammonia was introduced into the chamber. Within 24 hours of introducing the ammonia, the nicotine concentrations in the chamber increased to 4.5 μg/m^3. This result indicates that acid-base chemistry did affect the partitioning of nicotine between the sorbed phase on the carpet and the gas phase.

Results from experiments conducted in the large chamber are presented in Figures 3 and 4. In the first set of experiments, the chamber walls and ceiling were lined with painted gypsum wallboard, the floor was lined with nylon carpet, and the room was fully furnished (FF) as described previously. Background samples were collected before introducing ammonia into the chamber, and in all cases nicotine concentrations were ~ 1 μg/m^3 and ammonia concentrations were less than 1 ppm. In both fully furnished experiments, the ammonia concentrations peaked at ~ 11 ppm, 60-90 minutes after introducing the ammonia solution into the chamber. In the first furnished experiment, nicotine concentrations peaked at 45 μg/m^3, while in the second experiment nicotine levels rose to 14 μg/m^3 inside the chamber. The lower nicotine concentrations measured in the repeat experiment might be explained by considering that only a small fraction of the total adsorbed nicotine may have been available at the surface to be liberated by acid-base interactions. Much of this available nicotine might

have been re-emitted during the first experiment resulting in lower peak concentrations during the second trial.

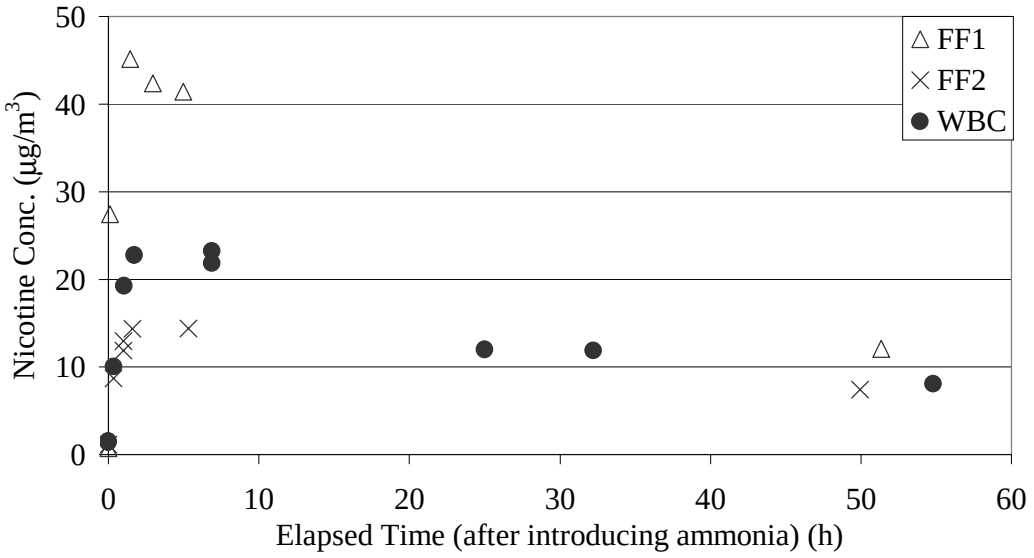

Figure 3. Nicotine concentrations in large chamber

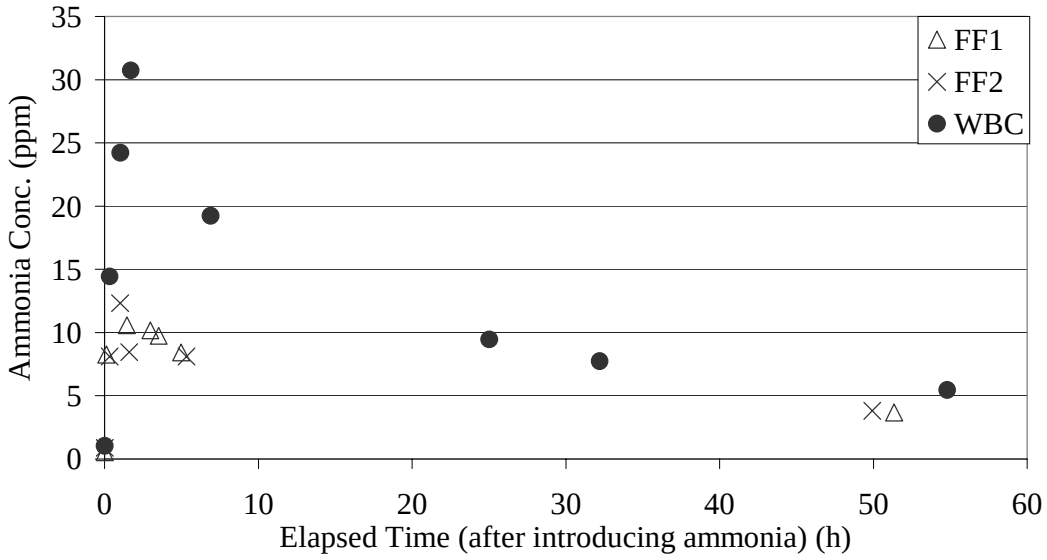

Figure 4. Ammonia concentrations in large chamber

In the final experiment, all of the furnishings were removed from the large chamber leaving only the wallboard and carpet (WBC). During this experiment, the ammonia concentrations peaked at 30 ppm approximately two hours after introducing the ammonia solution into the chamber. This peak concentration is three times higher than the peak observed in the previous two experiments. A possible explanation for this increase is that during the fully furnished experiments a large fraction of the ammonia was sorbing to the furniture, resulting in reduced airborne ammonia concentrations. The nicotine concentrations during the third experiment peaked at 23 $\mu g/m^3$, which is between the peak values for the first two rounds. This may be because elevated ammonia levels in the chamber resulted in an increased nicotine reemission rate from what might otherwise have been observed at lower ammonia concentrations.

CONCLUSION

These experiments have demonstrated that acid-base chemistry on surfaces can play a potentially significant role in altering indoor concentrations of certain pollutants. To better characterize such processes, it is important to consider the physical properties of the surface materials as well as the chemical properties of the pollutants of interest. The results of these specific experiments have shown that nicotine sorbs strongly to both wallboard and carpet. Increased reemission due to interactions with gaseous ammonia appears to be significant for carpet but not for wallboard. Common furnishings appear to provide additional sites for reversible nicotine sorption that is subject to gaseous ammonia effects. These experiments provide a basis for further study that could examine the effects of different chemicals, surface materials, relative humidity, and pH. Improving our understanding of the role of acid-base surface chemistry could aid in the development of more accurate sorption models as well as help determine how interactions of this nature may influence indoor air quality.

ACKNOWLEDGEMENTS

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MEASUREMENTS AND MODELING OF ENVIRONMENTAL TOBACCO SMOKE LEAKAGE FROM A SIMULATED SMOKING ROOM

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ABSTRACT
The purpose of this study is to quantify the effect of various design and operating parameters on smoking room performance. Twenty-eight experiments were conducted in a simulated smoking room with a smoking machine and an automatic door opener. Measurements were made of air flows, pressures, temperatures, two particle-phase ETS tracers, two gas-phase ETS tracers, and sulfur hexafluoride. Quantification of leakage flows, the effect of these leaks on smoking room performance and non-smoker exposure, and the relative importance of each leakage mechanism are presented. The results indicate that the first priority for an effective smoking room is to depressurize it with respect to adjoining non-smoking areas. Another important ETS leakage mechanism is the pumping action of the smoking room door. Substituting a sliding door for a standard swing-type door reduced this source of ETS leakage significantly. Measured results correlated well with model predictions (R^2 = 0.82-0.99).

INDEX TERMS
Environmental tobacco smoke, Ventilation, Chamber studies, Nicotine, Smoking room.

INTRODUCTION
The objective of this work was to quantify environmental tobacco smoke (ETS) leakage as a function of various smoking room operating and design parameters and measure its impact on performance. Smoking room performance was measured by releasing sulfur hexafluoride (SF_6) in a manner that simulated ETS generation. In some cases, however, the dynamics and transport of the various ETS components can differ substantially from that of SF_6 (Alevantis *et al.*, 1994) and from each other (Daisey, 1999). To address this issue, four particle- and gas-phase ETS tracers were measured in a subset of the tests.

Three potential ETS leakage mechanisms were investigated: (a) leakage through wall cracks or the gap under the door when the smoking room is pressurized relative to the non-smoking area; (b) leakage via the pumping action of the door as occupants enter and leave the smoking room; and (c) leakage through the overhead ceiling plenum. If the plenum above the smoking room is not isolated from the adjoining space's plenum, ETS can leak into the shared plenum. If ventilation systems for the non-smoking areas draw return air from the plenum, ETS can be recirculated into these areas. Keeping the smoking room depressurized relative to the overhead plenum will minimize leakage when the door is closed. Whenever the door opens, however, the smoking room pressure will quickly equilibrate with that of the non-smoking area and become higher than that of the plenum. This situation, while improved, can still result in sporadic bursts of ETS into the overhead plenum.

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The impact of these leakage mechanisms on smoking room effectiveness was assessed using two performance measures: smoking room exhaust efficiency and protection factor. The smoking room exhaust efficiency is the percentage of smoking room ETS that is successfully removed by the exhaust to the outdoors. The remainder can be assumed to escape from the smoking room into the surrounding, non-smoking areas. The steady-state exhaust efficiency, $\eta_{exh,}$ is given by

$$\eta_{exh} = Q_{exh,\,SR}\,[ETS]_{SR} \;/\; S \quad \text{x} \quad 100\% \qquad (1)$$

where $Q_{exh,\,SR}$ is the smoking room exhaust flow, $[ETS]_{SR}$ is the ETS concentration in the smoking room exhaust duct at steady-state, and S is the generation rate of ETS. In these experiments, SF_6 was released into the smoking room alongside the smoked cigarettes. SF_6 was injected at a known value of S and was thus a convenient, inert tracer for ETS leakage.

We have devised a new parameter, the smoking room protection factor (*SRPF*). *SRPF* represents the reduction in ETS concentration in non-smoking areas relative to the hypothetical case with no smoking room protection:

$$SRPF = [ETS]_{NSR,\,no\,SR} / [ETS]_{NSR} = (S / Q_{out,\,NSR}) / [ETS]_{NSR} \qquad (2)$$

where $[ETS]_{NSR}$ is the measured non-smoking room ETS concentration when a smoking room is used, $[ETS]_{NSR,\,no\,SR}$ is the non-smoking room ETS concentration that would have resulted from not using a smoking room (*i.e.*, smoking in the same space as the non-smokers), and $Q_{out,\,NSR}$ is the total flow out of the non-smoking area. For example, a measured *SRPF* value of 20 means that ETS concentrations in the non-smoking area are 20 times lower than they would have been if a smoking room had not been in operation. SF_6 was used as a tracer for ETS when calculating *SRPF* values.

Two particle-phase ETS tracers were measured: total particulate matter (PM) concentration and optical absorption of PM at 370nm (UVPM). UVPM has been found to be a sensitive and unique tracer of ETS (Gundel *et al.*, 2000). Two gas-phase ETS tracers were measured: nicotine and 3-EP.

METHODS

An existing chamber at Lawrence Berkeley National Laboratory (LBNL) was modified so that it consisted of two rooms separated by a wall with a door. The gap under the door was 0.64 cm. The two rooms were designated as the smoking room (SR) and non-smoking room (NSR), respectively. A suspended ceiling created a shared, 22.9 cm-high plenum above both rooms. The ceiling panels were of a common, “slag wool” (synthetic vitreous fiber) variety, and were cut to fit the lattice so that no gaps were visible. For the experiments with no shared plenum, the ceiling panels were removed and the plenum spaces above the two rooms were separated with a silicone-rubber-sealed acrylic divider.

Each room had a separate HVAC system. The supply air for each room passed through a high-efficiency filter, chiller coil, temperature-controlled duct heater, and a diffuser grille. SR air was exhausted to the outside. A fraction of the NSR exhaust airstream was returned to the NSR supply via a recirculation fan, and the remainder was exhausted to the outside. By adjusting the recirculation, supply, and return flows with regulating valves, a wide variety of pressure differences could be created between the SR, NSR, and ceiling plenum. The SR was equipped with an automated, programmable smoking machine built at LBNL. The smoking machine

could smoke 16 cigarettes consecutively. One cigarette was smoked at a time. A computer system controlled the smoking machine, a pneumatic-piston-based door-opening mechanism, and four rotating mixing fans. A door "open/close" cycle was initiated immediately before a cigarette was smoked and immediately afterwards, simulating a smoker entering and leaving the SR. SF_6 was released adjacent to the smoking machine.

Duct air flow rates, temperatures, and pressures were monitored using a data acquisition system. SF_6 was measured at 9 locations using 2 gas chromatographs with electron capture detectors and automated, multiport samplers. PM concentrations were determined gravimetrically using filter cassettes. ASTM Method #D5075-96 (ASTM, 1998) was used to determine gas-phase nicotine and 3-EP concentrations. Two dual-channel aethalometers were used to monitor UVPM in the NSR and NSR supply air.

Unless noted otherwise, each test typically lasted 4.5 hours, with 1.5 hours allowed for achieving steady-state chamber concentrations. The real-time samplers were used over the entire 4.5 hours, while the integrated samplers were operated for the last 3 hours of steady-state conditions only. Four sets of experiments were performed:

1) Leakage under closed door. These tests were conducted with the door closed and with no shared overhead plenum. Three positive values of the pressure gradient between SR and NSR were investigated, $\Delta P_{SR} = P_{SR} - P_{NSR} = 0$, 2.5, and 5 Pa.
2) Leakage via door pumping. These experiments lasted less than an hour and did not use a shared overhead plenum. The gap under the door was filled, the ventilation system was turned off, and $\Delta P_{SR} = 0$. In each test, SF_6 was allowed to build up in the SR with the door closed. After the injector was turned off, one door open/close cycle was performed, causing a burst of SF_6 to be pumped into the NSR. Tests were conducted using three different temperature gradients between rooms, $\Delta T_{SR} = T_{SR} - T_{NSR} = 2$, 0, and -2 ^{o}C. In addition to three tests with a swing-type door, one test was performed with a sliding door.
3) Leakage under door and via door pumping. In these tests, a swing-type door was opened on a regular schedule but was closed the remainder of the time, so ETS could leak both under the door and via door pumping. The major variable of these tests was ΔP_{SR} , which ranged from -10 to +5 Pa. These tests were performed using two different door-opening rates: 8 and 13.3 door cycles per hour (corresponding to 4 and 6.67 cigarettes per hour). In addition, two different ΔT_{SR} values were investigated: 0 ^{o}C and 2 ^{o}C.
4) Leakage through shared ceiling plenum. These tests were performed with the overhead plenum open between the SR and NSR and minimal pressure difference between rooms. All but one of these tests were performed with the door closed. Four positive values of pressure difference between SR and ceiling plenum were investigated, $\Delta P_{cp} = P_{SR} - P_{cp} = 0$, 0.3, 0.8, and 2 Pa. In addition, the effect of varying the SR exhaust airflow at one of these plenum pressures was studied. Finally, one test was performed in which the door opened 8 times/hour. During this test, the SR pressure was only higher than the plenum when the door opened.

One additional test was performed to investigate, within the limits of the test facility, the performance of a "no door" smoking room with high ventilation. In all, 27 experiments plus 5 replicates were performed. Cigarettes were smoked and ETS tracers were sampled in 9 of these tests plus 2 of the duplicates. Ventilation flow rates ranged from 0-100 L/s, corresponding to a ventilation range of 0- 14.6 ACH and 0-9.8 L/s/(m^2 floor area).

RESULTS

Smoking room leakage flows were determined for each experiment set using SF_6 mass balances. A least squares fit to the data from Experiment Set 1 produced the equation:

$$Q_{under\ door} = 6.10\ (\Delta P_{SR})^{0.573} \quad (3)$$

where $Q_{under\ door}$ is the flow under the door in units of L/s and ΔP_{SR} is in Pa.

The volume of air pumped by the door, $V_{door\ pump}$, was determined in Experiment Set 2. The average $V_{door\ pump}$ for the three swing-type-door tests was 672 L, with no apparent dependence on ΔT_{SR}. For the sliding door, $V_{door\ pump}$ = 152 L. The equivalent $Q_{door\ pump}$ is then

$$Q_{door\ pump} = D\ V_{door\ pump} \quad (4)$$

where D = number of door cycles / time.

A least squares fit to the data from Experiment Set 4 produced the equation

$$Q_{SR\text{-}cp} = 28.5\ (\Delta P_{cp})^{0.484} \quad (5)$$

where $Q_{SR\text{-}cp}$ is the flow from the SR into the ceiling plenum in units of L/s and ΔP_{cp} is in Pa. Then, the effective flow for a plenum leak that occurs only when the door opens is

$$Q_{SR\text{-}cp,\ open\ door\ only} = D\tau_{door}\ Q_{SR\text{-}cp} \quad (6)$$

where τ_{door} is the average time the door is open per cycle.

The leakage flows calculated with Equations 3 - 6 are plotted together on Figure 1 as a function of the pressure drop across the appropriate boundary. The plot assumes a swing-type

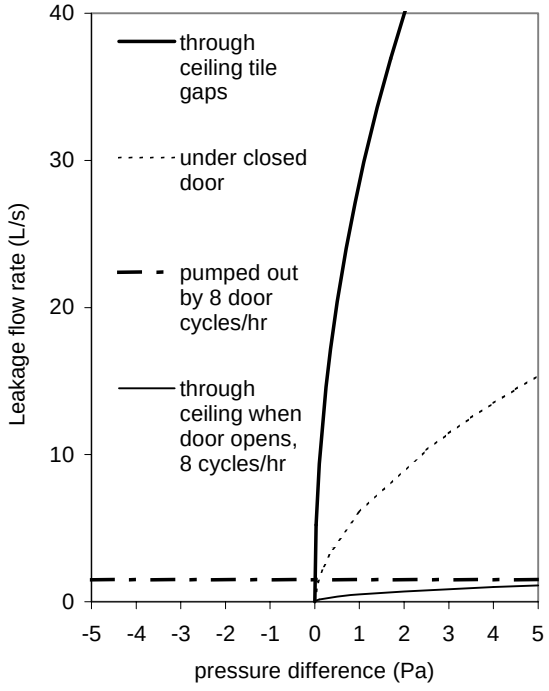

Figure 1. Comparison of SR leakage flows.

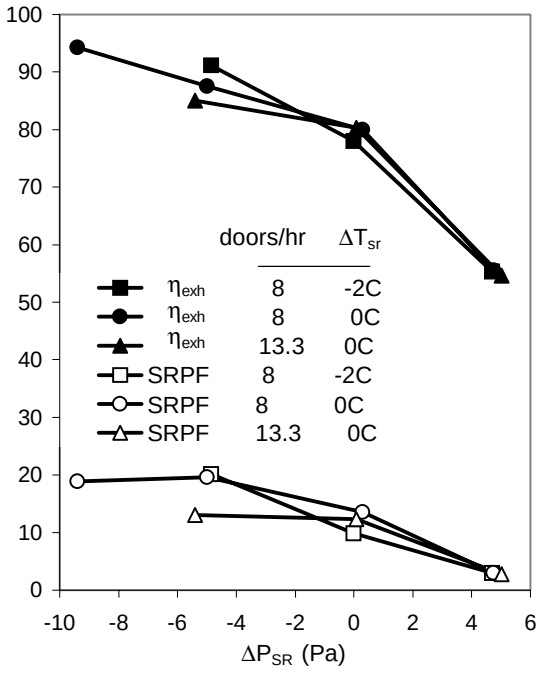

Figure 2. SR performance vs. pressure gradient.

door, D = 8 door cycles/hour and 8s/door opening. Assuming that ΔP_{SR} and ΔP_{cp} typically range within the same order of magnitude as each other, this plot can be used to determine the most important leakage mechanism in a given situation.

Steady-state η_{exh} and *SRPF* values were determined using Equations 1 and 2. Measured η_{exh} values ranged from 0-94%. Measured *SRPF* values ranged from 1.2 – 98. Figure 2 is a plot of η_{exh} and *SRPF* values measured in Experiment Set 3 as a function of ΔP_{SR}. Curves are plotted for ΔT_{SR} = 0 and –2 ^{o}C at (8 door cycles per hour) and for D = 13.3 door cycles per hour (ΔT_{SR} = 0). The η_{exh} and *SRPF* curves in Figure 2 correspond to a SR exhaust rate of 26.5 L/s. These results can be generalized to different building sizes and non-smoking-area ventilation rates, as η_{exh} and *SRPF* do not depend upon $Q_{exh,\,NSR}$. Figure 2 shows that both η_{exh} and *SRPF* were generally high when the SR was depressurized relative to the NSR, but decreased sharply as it became pressurized. Figure 2 also reveals that a temperature difference of 2 ^{o}C did not produce a measurable effect. Increasing the number of door cycles per hour did decrease η_{exh} and *SRPF*, but only when the SR was depressurized and door pumping was the dominant mechanism. Experiment Set 4 showed similar trends, with η_{exh} and *SRPF* decreasing as the SR became more pressurized with respect to the plenum. Increasing the SR exhaust flow rate was found to cause substantial improvements in η_{exh} and *SRPF*. Although increased SR exhaust flow does not prevent the various ETS leakage mechanisms from occurring, it does lower the ETS concentration in the SR, effectively reducing the strength of the leakage source.

Mass balances were performed for 23 of the tests to model η_{exh} and *SRPF* as a function of chamber pressures and exhaust flows. Correlation between modeled and measured results was high (R^2 = 0.99 and 0.83 for η_{exh} and *SRPF*, respectively).

In the NSR, correlations between the four ETS tracers and SF_6 ranged from 0.79 – 0.99. In the SR, the correlations ranged from 0.06 – 0.83. Exposure ratios (= $[ETS]_{NSR}/[ETS]_{SR}$) were calculated for each tracer as well. On average, exposure ratios calculated with 3-EP, PM, and nicotine were 34%, 41%, and 91% lower than those calculated with SF_6, respectively.

DISCUSSION

Equations 3 - 6 are useful for predicting SR leakage flows in actual buildings, as the inputs are relatively easy to obtain. The ΔP exponents in Equations 3 and 5 are both nearly equal to 0.5, which agrees well with the standard relationship for flow through an orifice. It should be noted, however, that these equations would have different coefficients for smoking room doors and ceiling panel configurations substantially different than the ones tested. For example, replacing some of the SR ceiling panels with grilles increased the leakage area substantially. These increases simultaneously reduced the pressure drop across the ceiling and increased the leakage flow. As a result, a suspended ceiling with several panels missing would likely have a very low pressure drop across the ceiling, but a substantially higher leakage flow than that predicted by Equation 5. Gaps under real-world smoking room doors vary as well, though to a lesser degree.

The pumping action when the door opens may have an enhanced impact on occupants of spaces immediately adjacent to the smoking room. The $V_{door\ pump}$ measured here for the swing-type door was 38% of the volume swept by door as it opened and closed. This value compares reasonably well the work of Kiel and Wilson (1989), who reported typical values of about 50%. The use of a sliding door instead of a conventional door is a promising method to reduce this

leakage mechanism. In the tested configuration, using the sliding door reduced ETS leakage via door pumping by 77%. Hypothetically, the use of a SR with no door could completely eliminate this ETS leakage source. However, thermally-induced circulations in our "no door" test caused NSR ETS levels to be not much lower than the case with no smoking room at all (*SRPF* = 2.4). Leakage through the ceiling plenum that occurs only when the door opens was observed to be a relatively minor leakage mechanism. Nevertheless, this mechanism can be eliminated by isolating the SR from the non-smoking area's ceiling plenum with a sealed wall.

Correlation between SF_6 and the ETS tracers was very good in the NSR, but poor in the SR. High nicotine and 3-EP levels in the SR likely caused significant sorption onto SR walls. Re-emission of these compounds on subsequent experiment days probably led to elevated air concentrations, even during experiments when PM and SF_6 emissions were low. Despite generally good correlation, the ETS tracers exhibited lower exposure ratios than SF_6, implying lower leakage to the NSR. Reduced PM exposure ratios may be partially due to differing semivolatile particle evaporation rates from the SR and NSR filters, which used two different sampling flow rates. Low nicotine exposure ratios are likely due to sorption of nicotine onto SR and NSR surfaces. The higher 3-EP exposure ratios are consistent with this interpretation, as 3-EP is more volatile and less sorptive than nicotine.

CONCLUSION AND IMPLICATIONS

ETS leakage flows have been quantified as a function of various operating and design parameters in a controlled chamber. Measured results correlated well with results modeled with simple mass-balance equations. This information can provide guidance for effective smoking room design and operation. The results indicate that the first priority for an effective smoking room is to depressurize it with respect to adjoining non-smoking areas. If this goal is achieved, the next most significant ETS leakage mechanism is the pumping action of the smoking room door when it is opened and closed. Substituting a sliding door for a swing-type door reduced this source of ETS leakage by 77%. The "no door" configuration tested in this study resulted in only modest non-smoker protection.

Although the relative importance of the various leakage mechanisms was determined with SF_6, correlation between the ETS tracers and SF_6 was generally good ($R^2 > 0.8$ in the NSR). Thus, these conclusions should apply to leakage of the ETS tracers as well. However, the magnitude of all the leakage mechanisms would be somewhat less for 3-EP and PM and substantially less for nicotine.

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IV. EXPOSURE, HEALTH AND PRODUCTIVITY STUDIES

INDOOR CARBON DIOXIDE CONCENTRATIONS AND SICK BUILDING SYNDROME SYMPTOMS IN THE BASE STUDY REVISITED: ANALYSES OF THE 100 BUILDING DATASET

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ABSTRACT
In previously published analyses of the 41-building 1994-1996 USEPA Building Assessment Survey and Evaluation (BASE) dataset, higher workday time-averaged indoor minus outdoor CO_2 concentrations (dCO_2) were associated with increased prevalence of certain mucous membrane and lower respiratory sick building syndrome (SBS) symptoms, even at peak dCO_2 concentrations below 1,000 ppm. For this paper, similar analyses were performed using the larger 100-building 1994-1998 BASE dataset. Multivariate logistic regression analyses quantified the associations between dCO_2 and the SBS symptoms, adjusting for age, sex, smoking status, presence of carpet in workspace, thermal exposure, relative humidity, and a marker for entrained automobile exhaust. Adjusted dCO_2 prevalence odds ratios for sore throat and wheeze were 1.17 and 1.20 per 100-ppm increase in dCO_2 ($p < 0.05$), respectively. These new analyses generally support our prior findings. Regional differences in climate, building design, and operation may account for some of the differences observed in analyses of the two datasets.

INDEX TERMS
Sick building syndrome, Ventilation, Carbon dioxide, Logistic regression, BASE study.

INTRODUCTION
Understanding the multifactorial etiology of sick building syndrome (SBS) in office buildings has been a major challenge. SBS is used to describe a set of symptoms with unidentified etiology frequently reported by workers in office buildings. The individuals who suffer from SBS report that the symptoms occur when they spend time indoors, particularly in office buildings, and that the symptoms lessen while away from the building (Levin, 1989; Mendell, 1993). Evidence for the hypothesis that building characteristics and resultant indoor environmental quality affects health outcomes continues to accumulate (Mendell, 1993; Fisk, 2000). These health outcomes include SBS symptoms, allergy and asthma symptoms, and respiratory illnesses. Indoor air quality also appears to influence rates of absence, work performance, and health care costs (Fisk, 2000). In this paper, we concentrate on building-related upper respiratory and mucous membrane (MM) symptoms (i.e., irritated eyes, throat, nose, or sinus), and lower respiratory (LResp) irritation (i.e., difficulty breathing, tight chest, cough, or wheeze).

The primary source of CO_2 in office buildings is respiration of the building occupants. CO_2 concentrations in office buildings typically range from 350 to 2,500 ppm (Seppänen et al., 1999). At concentrations occurring in most indoor environments, CO_2 buildup can be

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considered as a surrogate for other occupant-generated pollutants, particularly bioeffluents, and for ventilation rate per occupant, but not as a causal factor in human health responses. The Threshold Limit Value for 8-hour time-weighted-average exposures to CO_2 is 5,000 ppm (ACGIH, 1991). Currently, the American Society of Heating, Refrigeration, and Air-conditioning Engineers (ASHRAE) recommends a minimum office building ventilation rate offices of 10 Ls^{-1} per person, corresponding to an approximate steady state indoor concentration of 870 ppm (ASHRAE, 1999), based on the assumptions that outdoor CO_2 is 350 ppm and indoor CO_2 generation rate is 0.31 $Lmin^{-1}$ per person.

CO_2 And SBS Studies in the Literature

In a recent review (Seppänen et al., 1999), about one-half of 22 studies of SBS symptoms in office buildings found that increased indoor CO_2 levels were positively associated with a statistically significant increase in the prevalence of one or more SBS symptoms. SBS symptoms associated with CO_2 included headache, fatigue, eye symptoms, nasal symptoms, respiratory tract symptoms, and total symptom scores. Seventy percent of studies of mechanically ventilated and air conditioned buildings found a significant association between an increase in CO_2 and SBS symptoms. Building ventilation rates were also associated with SBS symptoms. An analysis of the 94-96 BASE dataset found statistically significant dose-response relationships between dCO_2 and the following symptoms: sore throat, irritated nose/sinus, combined mucous membrane symptoms, tight chest, and wheeze; the adjusted odds ratios for these symptoms ranged from 1.2 to 1.5 per 100 ppm increase in dCO_2 (Apte et al., 2000).

METHODS

The BASE Study

The data analyzed in this paper were collected in 100 randomly selected large U.S office buildings from 1994 to 1998 by the U.S. Environmental Protection Agency for the Building Assessment Survey and Evaluation (BASE) study (Girman et al., 1995; Womble et al., 1996). These buildings were at least partially mechanically ventilated and air conditioned. BASE buildings were studied during one-week periods either in winter or summer. Environmental data were measured during the week of questionnaire administration. The BASE protocol is discussed fully elsewhere (Womble et al., 1993; BASE Website).

The BASE questionnaire confidentially collected occupant information, including sex, age, smoking status, job characteristics, perceptions about the indoor environment, and health and well-being. The SBS symptoms elicited from the questionnaire included: irritation of eyes, nose, and throat; chest tightness, difficulty breathing, cough or wheezing; fatigue; headache; eyestrain; and dry or itchy skin. To qualify as a SBS symptom in the analyses presented here, the occupant must have had a reported a symptom occurrence at least 1-3 days per week during the month previous to the study and the particular symptom must have shown improvement when the occupant was away from work.

At each office building, CO_2, volatile organic compounds (VOCs), temperature, and relative humidity (RH) were measured at three indoor locations and outdoors. CO_2 and indoor temperature were collected as 5-minute averages. VOC canister samples were collected and analyzed by gas chromatograph-mass spectrometry for 73 VOC species. Spatial-average pollutant concentrations and average temperatures were calculated based on data from the three measurement sites. Time-averaged (8 hr) workday difference between indoor and outdoor CO_2 concentrations (dCO_2) was calculated as a surrogate measure of ventilation rate per occupant.

A thermal exposure variable (°C-hours) was calculated as the integrated difference between 5-minute-average-temperature and 20°C, normalized to 10 hours of exposure. The indoor workday-average relative humidity (RH) was calculated. The sixteen buildings with RH < 20% were excluded from the analyses discussed below, since by definition MM or LResp symptoms due to very low RH are not be considered SBS symptoms (Mendell, 1993).

Associations between BASE VOCs and SBS symptoms have been discussed elsewhere (Apte and Daisey, 1999). One VOC, 1,2,4 trimethylbenzene (TMB), found in infiltrating outdoor air and originating from automotive sources, was found to have statistically significant associations with a number of MM and LResp. Other sources of TMB in office buildings may include carpet, undercarpet, and building materials (Apte and Daisey, 1999). TMB was selected as a covariate in the regression models in order to adjust for the potential affects of ambient automotive sources on the SBS symptoms. The geometric mean TMB concentration across the 100 buildings was 0.6 ppb and the geometric standard deviation was 2.5.

Statistical Methods

Multivariate logistic regression (MLR) was used to calculate prevalence odds ratios (OR) and Wald Maximum Likelihood (WML) statistics (SAS, 1989). Crude and adjusted MLR models were constructed using continuous dCO_2 data as an independent variable and an SBS symptom as the dependent variable. Covariates used in the MLR models to control for confounding were age, sex, presence of carpet in workspace, smoking status, thermal exposure, RH, and TMB. Details regarding model building can be found in Apte et al. (2000). Additionally, a "California Buildings" covariate was added to some models.

RESULTS

Table 1 presents the results from both the crude and adjusted logistic regression analyses. The dCO_2 ORs are reported in units per 100 ppm. The larger 94-98 BASE data analysis yielded similar findings as compared with the smaller 94-96 data set, with smaller adjusted ORs ranging from 1.1 to 1.2 per 100 ppm increase in dCO_2 for Sore Throat, Nose/Sinus, and Wheeze. The effect for dry eyes observed in the smaller 94-96 dataset was not apparent in the larger 94-98 dataset.

Table 1. Crude and adjusted prevalence odds ratios[a] (OR) for dCO_2 and selected MM and LResp SBS symptoms for both the 94-96 and 94-98 BASE dataset analyses.

	94-96 BASE Dataset		94-98 BASE Dataset	
SBS	dCO_2 OR (per 100 ppm)		dCO_2 OR (per 100 ppm)	
Symptom	Crude	Adjusted	Crude	Adjusted
MM				
Dry eyes	**1.1 (1.04-1.23)**[b]	**1.2 (1.06-1.29)**[b]	1.0 (0.99-1.11)	1.0 (0.98-1.12)
Sore throat	**1.4 (1.21-1.59)**[b]	**1.5 (1.25-1.72)**[b]	**1.2 (1.09-1.31)**[b]	**1.2 (1.06-1.29)**[b]
Nose/sinus	**1.1 (1.04-1.26)**	**1.2 (1.06-1.34)**[b]	1.1 (0.98-1.14)	1.1 (0.99-1.15)
LResp				
Chest tight	1.1 (0.90-1.41)	1.3 (0.96-1.66)	1.0 (0.85-1.19)	1.0 (0.86-1.21)
Short breath	1.1 (0.87-1.37)	1.3 (0.97-1.69)	1.0 (0.87-1.24)	1.1 (0.92-1.35)
Cough	1.1 (0.91-1.23)	1.1 (0.90-1.28)	1.0 (0.86-1.07)	0.95 (0.85-1.07)
Wheeze	**1.4 (1.14-1.78)**[b]	**1.4 (1.07-1.84)**	**1.2 (1.02-1.42)**	**1.2 (1.00-1.42)**

[a]Values in parentheses are the 95% confidence interval (CI). ORs and CIs given in bold are statistically significant at the 95% confidence level or higher.

[b] $p \leq 0.005$

Preliminary analyses investigated why the results obtained from the larger 94-98 dataset differed from those obtained from the smaller 94-96 dataset. Mean levels and standard deviations of dCO_2 and the continuous covariates did not differ substantially between buildings for which data were collected in 94-96 compared with buildings for which data were collected more recently (see Table 2). Of the dichotomous covariates, only the proportion of females and older occupants differed between the two data collection periods (see Table 3). In terms of SBS symptom prevalences, the two data collection periods did not differ appreciably (see Table 3).

Table 2. Means and standard deviations for dCO_2 and continuous covariates.

	94-96 BASE Buildings		97-98 BASE Buildings		
Variable	Mean	SD	Mean	SD	P-value[a]
dCO_2 (ppm)	242	142	288	130	0.12
thermal exposure	26.16	6.84	24.37	6.94	0.25
RH	40.28	8.71	44.51	10.97	0.06
TMB	1.28	1.31	0.93	0.96	0.17

[a]Student's t-test, 2-sided

Table 3. Percent of occupants reporting selected characteristics and SBS symptoms.

Variable	94-96 BASE Buildings	97-98 BASE Buildings	P-value[a]
% female	68.0	64.8	0.04
% $\geq$ 40 years	53.2	57.5	0.01
% with carpet	89.9	90.5	0.50
% current smoker	15.5	14.0	0.24
% MM	27.9	26.3	0.30
% dry eyes	20.3	18.8	0.32
% sore throat	7.0	6.9	0.95
% nose/sinus	13.5	12.8	0.58
% LResp	8.8	7.7	0.29
% chest tight	2.4	2.2	0.72
% short breath	2.3	1.5	0.12
% cough	5.3	5.4	0.94
% wheeze	2.4	1.8	0.22

[a]Chi-square, Fisher's exact test, two-sided

Regional differences in climate, building codes, and other factors may account for the differences in the two analyses. For example, changes in indoor smoking policies over the BASE study period may have had an impact on factors that may be associated with SBS symptoms. One way to examine the influence of regional differences is to look at the influence of buildings from different states. Because California contributed the largest proportion of recently added buildings in 1997-1998 to the BASE study (16%), we first considered whether being in a "California Building" influenced SBS symptoms. After including the "California Building" covariate in the MLR model, the odds ratios relating dCO_2 (per 100 ppm) to the selected SBS symptoms more closely resembled those found in the analysis of the 94-96 dataset (see Table 4).

DISCUSSION

The results of these analyses indicate an association between elevated indoor CO_2 levels and increases in certain MM and LResp SBS symptoms. These findings were evident in the crude

regression models and persisted through adjustment for a number of potential confounders. After adjusting for whether a building was in California, the OR for combined MM and nose/sinus symptoms also achieved statistical significance, thus highlighting the potential importance of regional effects. Investigating specific regional differences provides an opportunity to identify building characteristics that are associated with better indoor air quality and lower SBS symptom prevalence. Subsequent analyses will employ more sophisticated models to explore the potential impact of cross-level bias.

Table 4. Adjusted prevalence odds ratios[a] (OR) for dCO_2 per 100ppm, the California building variable and selected MM and LResp SBS symptoms for the 94-98 BASE dataset analysis.

SBS Symptom	dCO_2 (per 100 ppm) OR	California Building[b] OR
MM	**1.1 (1.01-1.14)**	**1.3 (1.01-1.59)**
Dry eyes	1.1 (1.00-1.14)	1.2 (0.95-1.59)
Sore throat	**1.2 (1.09-1.35)**	1.5 (0.98-2.18)
Nose/sinus	**1.1 (1.02-1.19)**	**1.4 (1.07-1.92)**
LResp	1.1 (0.96-1.17)	1.3 (0.94-1.92)
Chest tight	1.1 (0.90-1.29)	1.8 (0.94-3.28)
Short breath	1.1 (0.91-1.38)	1.1 (0.48-2.37)
Cough	1.0 (0.85-1.09)	1.1 (0.72-1.74)
Wheeze	**1.3 (1.04-1.51)**	1.7 (0.80-3.42)

[a]Values in parentheses are the 95% confidence interval (CI). ORs and CIs given in bold are statistically significant at the 95% confidence level or higher.
[b]Estimating the association between being a "California Building" and SBS symptoms.

The odds ratios for the associations of symptoms with the maximum observed difference between indoor and outdoor CO_2 concentrations may indicate the maximum potential to reduce selected SBS symptoms in typical office buildings. The maximum value of dCO_2 was 608 ppm. Considering only the significant associations, the ORs for the maximum value of dCO_2 range from 6.7 to 7.3. Based on these ORs, the implied potential maximum reduction in prevalence of these symptoms is roughly 85%. This reduction could come through large increases in ventilation rates, improved effectiveness in providing fresh air to the occupants' breathing zone, or through identification of the symptom-causing agents in the indoor air and control of their sources. In no case were the indoor average or the peak indoor CO_2 concentrations extraordinarily high; only two buildings had peak indoor (absolute) CO_2 concentrations routinely above 1,000 ppm.

CONCLUSION AND IMPLICATIONS

After adjusting for selected covariates, we found statistically significant associations of mucous membrane and lower respiratory SBS symptoms with increasing dCO_2. Odds ratios for statistically significant associations of sore throat and wheeze symptoms with 100-ppm increases in dCO_2 were 1.1 to 1.2. These results suggest that increases in the ventilation rates per person among typical office buildings will, on average, significantly reduce the prevalence of several SBS symptoms, even when these buildings meet the existing ASHRAE ventilation standards for office buildings. The magnitude of the reduction will depend on the magnitude of the increase in ventilation rates, improvement in ventilation effectiveness, or reduction in sources of SBS-causing agents. Very large increases in ventilation rates, sufficient to reduce indoor CO_2 concentrations to approximately outdoor levels, would be expected, on average, to decrease prevalence of selected symptoms by 85%. There is no direct causal link between

exposure to CO_2 and SBS symptoms, but rather CO_2 is approximately correlated with other indoor pollutants that may cause SBS symptoms. The BASE dataset is a valuable source of U.S building information, providing an opportunity for identification of causal factors of SBS, and for development of solutions for lowering its prevalence in buildings.

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WORKER PERFORMANCE AND VENTILATION: ANALYSES OF INDIVIDUAL DATA FOR CALL-CENTER WORKERS

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ABSTRACT
We investigated the relationship between ventilation rates and work performance in a call center. We randomized the ventilation controls and measured ventilation rate, differential carbon dioxide (ΔCO_2) concentration, temperature, humidity, occupant density, degree of under-staffing, shift length, time of day, and time required to complete two different work performance tasks (talk and wrap-up). ΔCO_2 concentrations ranged from 13 to 611 ppm. We used multi-variable regression to model the association between the predictors and the responses. We found that agents performed talk tasks fastest when the ventilation rate was highest, but that the relationship between talk performance and ventilation was not monotone. We did not find a statistically significant association between wrap-up performance and ventilation. At high temperatures agents were slower at both the talk and wrap-up tasks. Agents were slower at wrap-up during long shifts and when the call center was under-staffed.

INDEX TERMS
Ventilation rates, Productivity, Offices, Worker performance, Carbon dioxide

INTRODUCTION
This paper describes part two of a two-part analysis from a productivity study performed in a call center operated by a health maintenance organization. The companion paper (Fisk et al., 2002) describes the analysis of group performance data that has good time resolution. The objective of this paper is to report the on the analysis of individual performance data acquired from the same experiment. The analysis described in this paper confirms the findings in the companion paper and reveals additional insight into the relationship between environmental factors and work performance.

METHODS
The study design and procedures are described in the companion paper (Fisk et al., 2002). In this section we describe methods that are specific to the analysis of the individual performance data.

The call center is an “inbound” call center, which means that calls originate from clients. Calls are forwarded to agents by an automated call distribution (ACD) system. The task of processing a call is referred to as “handling” a call. Handling a call involves two subtasks referred to as “talk” and “wrap-up.” The talk task involves the period of time when the agent is talking with the client. Wrap-up involves performing tasks such as database entry after the agent has finished talking with the client. When wrap-up is complete the agent signals the ACD system that he is ready to accept another call.

Two kinds of agents work in the call center, registered nurses (RNs) who provide medical advice and tele-service representatives (TSRs) who screen calls and schedule appointments. In this paper we only consider data related to the RNs.

The ACD system measures the duration of every talk task and every wrap-up task. Agents sign in to the ACD system with a unique identification number. We analyzed data from reports showing the average talk time and wrap-up time during each shift for each agent coded by the agent identification number. Individual performance per task or over shorter time intervals than a whole shift were not available. Our response variables were the natural logarithm of the shift-averaged talk times and wrap-up times.

Agents sign out of the ACD system when they finish a shift. We used reports with sign-in and sign-out times for each agent each day to determine the length of each agent's shift.

The ACD system records a metric called NETS. NETS is the number of agents scheduled minus the number needed to keep the waiting time in the queue equal to a target. When NETS is negative the call center is under-staffed.

The call center is organized into clusters of cubicles called "neighborhoods." Most agents work in the same neighborhood each shift. As reported in Fisk et al. (2002), we recorded space temperature and humidity continually during the study in each neighborhood. By associating agents with neighborhoods we were able to associate individual temperature and humidity readings with agents.

The call center has four air-handling units of approximately equal size. For each air-handling unit we recorded the difference between the carbon dioxide concentration in the return duct and the outdoors (ΔCO_2). We also determined the outdoor airflow rate based on supply airflow and CO_2 concentrations in the return, outdoor, and supply ducts. We associated the ΔCO_2 concentrations and outdoor airflow rates with individual agents by using floor plans to determine which neighborhoods each air-handling unit served.

We computed shift-averaged NETS, temperature, humidity, ΔCO_2, and ventilation rate for each worker. The averages were used as predictors in our models. We used multi-variable regression to model the association between the response variables and the predictors. The ACD system records the number of calls per shift per agent. We used the number of calls as weights in our regression analysis. Our models are multiplicative. The logarithm of the talk time or wrap-up time is modeled as a linear combination of functions of the response variables.

The call center operates 24 hours per day, 7 days per week. We only analyzed data associated with shifts starting after 6am and finishing before 10pm, Monday-Friday. We removed obvious outliers manually. The software that runs the ACD system was upgraded 9 days before the start of the study and on the 58^{th} day of the study (a Friday evening). We observed a transient in the daily-averaged wrap-up times during the first two weeks of the study, so we discarded the first day (a Friday) and the following two weeks of data. We found that the transient in the daily-average wrap-up times was shorter after the second software upgrade, so we only discarded one week of data after that event. We included an indicator variable to account for the performance difference after the second software upgrade.

RESULTS

Descriptive statistics for the explanatory variables are shown in Table 1. The ventilation rate distribution has three modes that correspond to low, medium, and high damper settings. The temperature distribution has a long high-temperature tail. The relative humidity has a long low-humidity tail. The number of occupants includes TSRs because occupant density is a proxy for auditory and visual distractions. The distribution of shift length has four modes corresponding to 4, 5, 6, and 8 hour shifts. Below the 4-hour mode there is a long tail of short shifts.

Table 1: Descriptive statistics of explanatory variables.

	ΔCO_2	Vent	Temp	RH	Occ.	NETS	Shift
min	13 ppm	0.26 l/s-m^2	21.0 C	20.2 %	66.6	-11	0.48 hrs
med	243	1.38	23.0	42.5	156.3	1.8	6.4
mean	253	2.07	23.1	42.4	159.6	2.7	6.8
max	611	10.0	26.6	55.3	253.6	17.2	12.4

We tried models with linear and categorical terms for ΔCO_2, a linear ΔCO_2 term multiplied by shift length, linear, inverse linear, and categorical terms for ventilation rate, ventilation rate multiplied by shift length, quadratic and categorical terms for temperature, quadratic terms for enthalpy, a linear term for occupant density, quadratic and categorical terms for NETS, and linear and categorical terms for shift length. The following results are for a model with a categorical term for either ΔCO_2 or ventilation rate, a categorical temperature term, a linear occupant density term, a categorical NETS term, and a categorical shift length term.

Figures 1 and 2 show the predicted association between talk time and the ΔCO_2 and ventilation categories. Positive "effects" mean the agents worked slower. In both cases, the highest ventilation rate (lowest $\Delta CO_{2)}$ is the baseline. Vertical bars in these and all successive figures are standard errors. Boundaries for the ΔCO_2 categories are at 100, 250, and 420 ppm. The ΔCO_2 p-values are 0.012, 0.15, and 0.60. The boundaries for the ventilation categories are 0.76, 2.4, and 4.4 l/s-m2. The ventilation rate p-values are 0.32, 0.047, and 0.022.

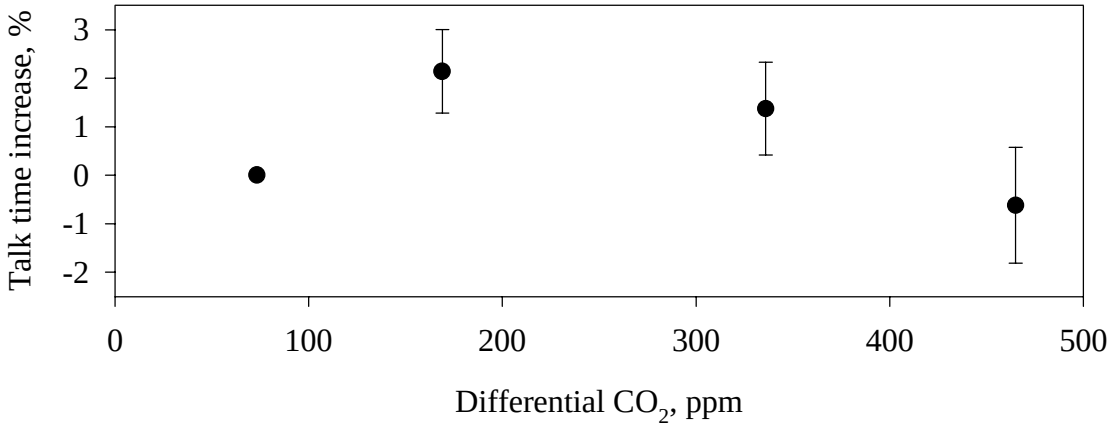

Figure 1: ΔCO_2 category coefficients for talk model.

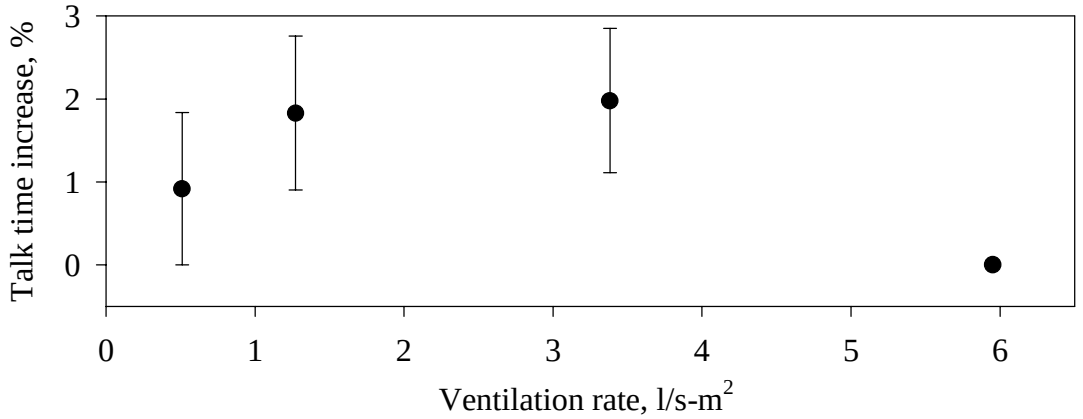

Figure 2: Ventilation category coefficients for talk model.

The model with ΔCO_2 categories predicts that agents take 8.5% longer to talk at maximum occupancy than at minimum occupancy (p=0.05). The model with ventilation categories predicts that they take 4% longer (p=0.36). The models predicts that agents performed the talk task 4.0% and 4.3% slower after the software upgrade (p=0).

The models of wrap-up time showed no statistically significant association between wrap-up time and differential ΔCO_2 or ventilation rate. They did show an association between wrap-up time and temperature, NETS, and shift length.

Figure 3 shows the coefficients of the temperature categories for the wrap-up model containing ΔCO_2 categorical variables. The baseline is 23 °C. The boundaries between the categories are 21.7, 22.5, 23.5, 24.5, and 25.4 °C. The p-values for the coefficients are 0.89, 0.26, 0.91, 0.98, and 0.0028. The model with ventilation rate categories also predicts lower performance at high temperature.

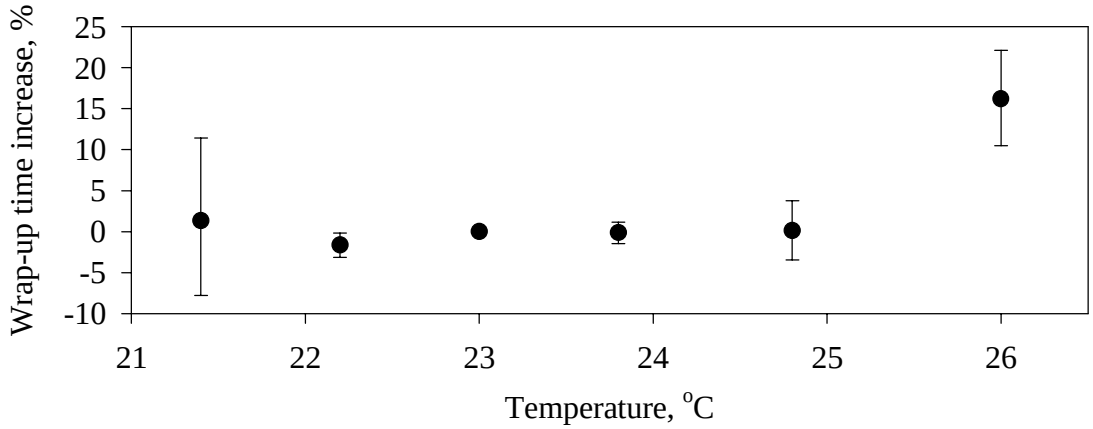

Figure 3: Temperature coefficients for wrap-up model with ΔCO_2.

Figure 4 shows the coefficients of the NETS categories for the same wrap-up model. The baseline is NETS=0, meaning no under-staffing or over-staffing. There is a clear trend showing that agents perform wrap-up fastest when NETS is positive (call center overstaffed). The p-values for the NETS coefficients are 0.046, 0.12, 0.30, 0.11, and 0.55.

Figure 5 shows the coefficients of the shift length categories for the same wrap-up model. There is a clear, increasing trend. All of the coefficients less than the baseline (nominal 8-hour shift) are less than zero while the coefficient greater than the baseline is greater than zero. The p-values for the coefficients are 0.060, 0.72, 0.57, 0.0070, and 0.23.

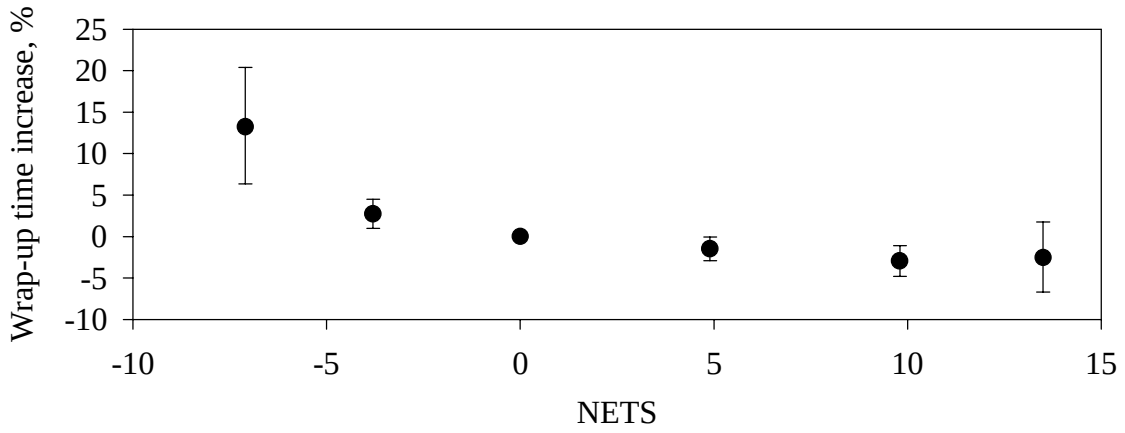

Figure 47: NETS coefficients for wrap-up model with ΔCO_2.

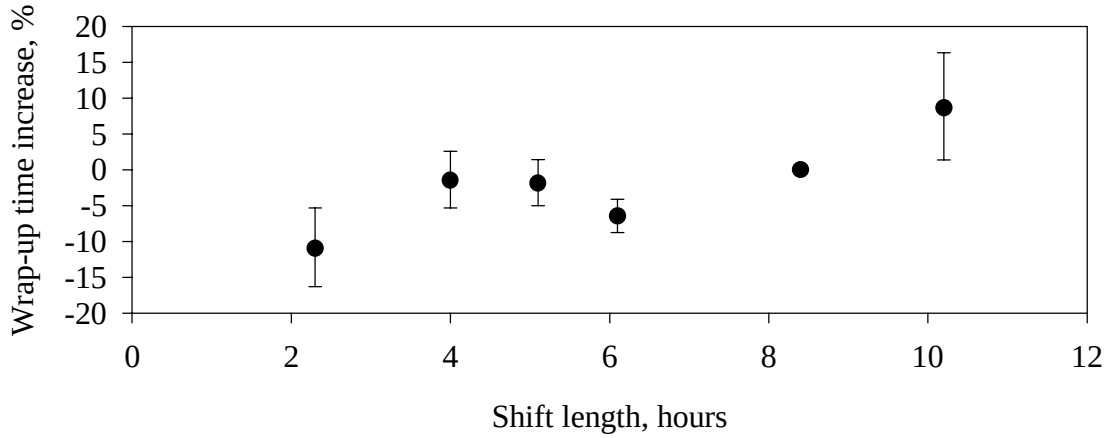

Figure 5: Shift length coefficients for wrap-up model with ΔCO_2.

The wrap-up model with ΔCO_2 categories predicts that wrap-up is 19.7% longer at full occupancy than at minimum occupancy (p=0.067). The same model predicts that wrap-up was 7.5% shorter after the software upgrade (p=0).

DISCUSSION

We expected, but did not find, a dose-response relationship between ventilation and work performance. Instead we found that performance at low and high ventilation rates was statistically the same, that performance at intermediate ventilation rates was worse by about 2%, at that the increase was statistically significant. We tried a number of different models, including models containing the air quality proxies multiplied by shift length (time of exposure). The pattern described above showed up in most of these models.

We found that what evidence there was of an association between ventilation and performance showed up in the talk task and not in the wrap-up task. We expected that

productivity loss would be more likely during wrap-up because agents are free to work at their own pace; they are not constrained by their interaction with a client.

We found that agents were 16% slower at wrap-up when the temperature was greater than 25.4 °C. This was the largest effect with a p-value less than 5%. However, temperatures greater than 25.4 °C occurred on only seven days, six of them in a nine-day period starting at day 21. Some models we tried showed a similar increase in wrap-up times at low temperatures, but the low-temperature terms were never statistically significant.

When the call center was understaffed agents performed wrap-up slower. The call center director predicted this effect prior to our analysis. The hypothesis is that when the call center is understaffed agents never get to rest between calls. They make up for the missing rest by working slower during wrap-up.

CONCLUSION AND IMPLICATIONS

1. There is some evidence that ventilation rates less than 100% outdoor air are associated with lower work performance, but the results are not conclusive.
2. There is some evidence that high temperature (> 25.4 C) is associated with lower work performance.
3. There is some evidence that higher occupant density is associated with lower work performance.
4. Understaffing is associated with lower work performance.
5. Longer shifts are associated with lower work performance.

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WORKER PERFORMANCE AND VENTILATION: ANALYSES OF TIME-SERIES DATA FOR A GROUP OF CALL-CENTER WORKERS

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ABSTRACT
We investigated the relationship of ventilation rates with the performance of advice nurses working in a call center. Ventilation rates were manipulated; temperatures, humidities, and CO_2 concentrations were monitored; and worker performance data, with 30-minute resolution, were collected. Multivariate linear regression was used to investigate the association of worker performance with building ventilation rate, or with indoor CO_2 concentration (which is related to ventilation rate per worker). Results suggest that the effect of ventilation rate on worker performance in this call center was very small (probably less than 1%) or nil, over most of the range of ventilation rate (roughly 12 L s^{-1} to 48 L s^{-1} per person). However, there is some evidence of worker performance improvements of 2% or more when the ventilation rate per person was very high, as indicated by the indoor CO_2 concentration exceeding the outdoor concentration by less than 75 ppm.

INDEX TERMS
Carbon dioxide, Offices, Productivity, Ventilation rates, Worker performance

INTRODUCTION
In previous studies, increased ventilation rates and reduced indoor carbon dioxide concentrations have been associated with improvements in health at work (Seppanen et al., 1999) and with increased performance in work-related tasks. Only a few studies have assessed the relationship of ventilation rates with worker performance. In a study of 35 Norwegian classrooms, higher concentrations of CO_2, which indicate lower rates of outside air ventilation per person, were associated with poorer performance ($p < 0.01$) in computerized tests of reaction time (Myhrvold et al., 1996); however, the percentage change in performance was not specified. In a study by Nunes et al. (1993), workers who reported building-related health symptoms, known to be associated with lower ventilation rates (Seppanen et. al., 1999), took 7% longer to respond in a computerized neurobehavioral test of sustained visual attention ($p < 0.001$) and had 30% higher error rates in a symbol-digit substitution test of speed and coding ability. In laboratory experiments by Wargocki et al. (2000), increasing the ventilation rate in a room with a carpet from a complaint building was associated with improvements of a few percent in speed or accuracy of several simulated work tasks such as text typing, addition, proof reading, and creative thinking ($p < 0.05$).

The difficulty of defining and measuring the cognitive performance of workers has been one of the barriers to studying worker performance in real work places. However, for a few types of cognitive work, worker performance has been clearly defined and routinely measured by the employer. For example, in call centers, large pools of workers interact with clients via the

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telephone and enter data or process information associated with the telephone calls. To track worker performance, call centers frequently have automatic systems that record data on worker speed and type or purpose of the calls. Consequently, call centers are an appropriate setting for studies of the dependence of work speed, but not work quality, on indoor environmental quality (IEQ). This paper describes such a study in a call center operated by a health maintenance organization.

METHODS

The approach employed in this study was to manipulate outside air ventilation rates and monitor indoor air temperatures (which varied naturally), while collecting telephone call data that quantified worker performance at a call center located in the San Francisco Bay Area. Data were collected between July 28 and October 24, 2000 and analyzed with multivariate statistical models. The workforce was blinded regarding all aspects of the study, except that they were aware that indoor air temperatures were being monitored.

The call center had a floor area of 4,600 m^2, sealed windows, carpeted floors, a no smoking policy, and a maximum worker density of 6.3 persons per 100 m^2. The call center was heated, cooled, and ventilated by variable air volume (VAV) air handling units (AHUs) that modulated the rates of supply of cool or warm air to maintain indoor air temperatures in the desired range. Each AHU had an "economizer" control system that modulated the rate of outside air supply, above a minimum rate established by the building code, with the goal of minimizing costs for heating and cooling; however, to prevent unplanned changes in outside air supply the economizer controls were deactivated during most experimental periods. The workers were registered nurses (RNs) who provided medical advice. Each RN had a computer and telephone with a headset. Workers were present in the building at all times and days, although the number of workers was highly variable, with the largest workforce on weekday mornings. The maximum number of RNs in the building during this study was 119.

We added equipment to each AHU enabling automatic manipulation and measurement of outside air ventilation rates. AHU supply flow rates were measured using arrays of pitot tubes in supply ducts, with the pressure differences logged. The outside air flow rate in each AHU was computed as the product of the supply airstream flow rate and the fraction of outside air (FOA) in this airstream. We used a CO_2 monitor calibrated weekly to measure concentrations of CO_2 every 7.2 minutes in the outside air, return, and supply airstreams and employed a simple mass balance calculation to compute the FOA.

The AHUs had dampers for modulation of the FOA. Fixed damper positions for low ventilation rate periods were selected to match the code-minimum outside air supply rate of 12.0 L s^{-1} per occupant at maximum occupancy (0.76 L s^{-1} per square meter of floor area and 292 persons). The fixed damper positions for medium and high ventilation rates were selected to provide approximately twice and four-times the code minimum. In a fourth ventilation setting, the normal control systems for the AHU's outside air supply, including the outside air economizers, were activated. We anticipated that this mode of operation (called economizer mode) would typically provide a ventilation rate greater than eight times the code minimum. In practice, ventilation rates in economizer mode varied considerably.

Using these methods, we scheduled periods of ventilation in each of the four control modes: low, medium, high, and economizer mode. The intent to have randomized ventilation rates that changed daily during weeks 3 – 6 and 8 – 10 was met reasonably well. During weeks 1, 2, 7, 11 and 13, we intended to fix the ventilation rates at the low, medium, high, or

economizer setting for one-week periods; however, the control system failed during some periods. The resulting schedule of ventilation control modes is provided in Table 1. The control system resulted in a wide range of ventilation rates; however, these ventilation rates were not sufficiently repeated to use the control mode as a categorical surrogate for the ventilation rate. Thus, measured ventilation rates and carbon dioxide concentration were used in analyses of the worker performance data.

Table 1. Ventilation control schedule. L, M, and H refer to fixed damper positions for low, medium, and high ventilation rates. E refers to control of ventilation rates by the economizer.

Day	**Week**												
	1	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**	**13**
F	L	H	E	H	L	E	M	M	H	M	E	E	E
Sa	L	H	L	E	H	M	M	M	M	L	E	E	E
Su	L	H	H	L	M	L	M	M	E	M	E	E	E
M	L	H	E	H	L	M	M	M	M	H	E	E	L
Tu	L	H	M	E	H	L	M	L	H	E	H	H	E
W	L	H	H	L	E	H	M	M	L	E	H	E	--
Th	L	H	M	H	M	E	M	L	E	E	H	E	--

Air temperatures were measured, with an accuracy of approximately 0.3 ^{o}C, every one minute at 25 locations approximately one meter above floor level.

The call center's automated call distribution (ACD) system monitored several performance-related parameters. Worker performance for each half-hour period was summarized with the "average handle time"(AHT) of all of the calls that ended during that period. The AHT is the average time (averaged over all RNs and over the entire half hour) taken for each call, starting when the call was answered and ending when the RN completely finished all tasks associated with the call. For each half-hour period, the call center's computer calculated a number, called "nets", that estimated (based on prior experience and current number of incoming calls) how many extra RNs were on hand, compared to the number needed to have the average wait experienced by callers equal to a target wait time. For periods when the target wait times were exceeded, nets was negative. Nets was used as a variable in the data analyses, as a measure of the work demand on RNs.

Our primary interest was the relationship between ventilation rate and RN performance measured by AHT. We expected ventilation rate to influence AHT by at most a few percent, which is less than the variation due to several other sources. To estimate effects of ventilation rate on performance with useful precision, we excluded some data from the analyses and controlled for other sources of variation in AHT using multivariate regression models. We discarded data outside the normal work week (Monday – Friday, 7:30 a.m. – 6:00 p.m.) when few workers were present. Data from a holiday (Labor Day) and the following day were excluded, and data from two additional days were excluded due to changes in computer software.

As a proxy for ventilation per agent, we used the difference between the indoor and outdoor concentration of CO_2 (ΔCO_2) [linear, categorical, or piecewise linear] as the explanatory variable of primary interest. Linear regression was our main tool for analyzing the data: we regressed log(AHT) on explanatory variables that were expected to be relevant. To control for potential confounding, we included a number of other variables in the regression models:

number of RNs present, average relative humidity, a time-of-week indicator variable for each half-hour period [which accounted for about 35% of the variation in log(AHT)], building-average temperature (Celsius) minus 23 ^{o}C , and (building-average temperature minus 23 ^{o}C)^2 to account for the possibility of a non-linear relationship between temperature and performance. We also included a piecewise linear normalized "nets" variable, normalized to the number of workers on duty. Each data point was weighted by the number of calls received during the half hour, although the weights were not highly variable, nor were they very influential. As described in Fisk et al (2001), the linear regressions adjusted for the temporal correlation of the residuals.

RESULTS

ΔCO_2 concentrations were rarely below 100 ppm or above 450 ppm. ΔCO_2 values tended to cluster into three wide clumps, corresponding to low, medium, and high damper settings, with "economizer" settings also tending to lead to fairly low or very low values of ΔCO_2.

The AHUs held the building-average temperature within a very narrow range during working hours. Ninety percent of the half-hourly work-day temperatures were between between 22.9 ^{o}C and 23.5 ^{o}C. Building average humidity almost never strayed outside the range 46% to 47%. Unsurprisingly, then, the regression coefficients associated with temperature variables and relative humidity, were all very small compared to their uncertainties.

We fit several dozen regression models, using different definitions for the bin boundaries for number of calls, normalized nets, and ΔCO_2, and using different subsets of the data. Measures of model fit were very similar for all models that included the full set of explanatory variables. In every model, the "nets" variables were highly influential.

We now discuss two specific models for log(AHT) in some detail. Each model includes the time-of-week, temperature, and "nets" variables. The two models differ in their handling of ΔCO_2. Model B includes three ΔCO_2 categorical variables, indicating whether ΔCO_2 for each half hour was: 0-150 ppm, 150-300 ppm, or over 300 ppm. In Model C, the two lower ΔCO_2 categories within Model B have been split, thus, Model C has five ΔCO_2 categorical variables: 0-75 ppm, 75-150 ppm, 150-225 ppm, 225-300 ppm, or over 300 ppm. Figure 1 shows the estimated model coefficients associated with each ΔCO_2 bin, for Models B (lower plot) and C (upper plot). For each bin, the horizontal bar shows the range of ΔCO_2 spanned by the bin, and the vertical error bar covers plus or minus one standard error. In each case, the lowest bin is defined to have no effect, a coefficient of 0.00.

In Model B with only three ΔCO_2 bins, there is no evidence that lower ΔCO_2 is associated with lower (faster) AHT -- indeed, the relationship points the other direction: the estimate for the high-ΔCO_2 bin is about 1% faster than that for the lowest bin. (An effect of -0.009 on log(AHT) corresponds to a factor of exp(-0.009)=0.991 on AHT, which is very close to a 1% speed-up). However, this estimate is not very precise, with an uncertainty (one standard error) of approximately ± 0.6 percentage points. In contrast, the results from Model C with five bins suggest that very low ΔCO_2 concentrations are associated with lower AHT (faster work) than are higher concentrations. All of the estimated coefficients for ΔCO_2 concentrations over 75 ppm are around 0.025 to 0.035, corresponding to handle times that are 2.5% to 3.5% slower than at the lowest ΔCO_2. Moreover, these effects are all statistically significant ($p < 0.05$ for all bin coefficients). However, as we discuss below, we think the relationship between AHT and ΔCO_2 is still far from conclusive.

Neither the Model B nor Model C results provide evidence that handle time increases with ΔCO_2 over most of its range. A dependence of log(AHT) on ΔCO_2 is apparent only for ΔCO_2 below about 150 ppm. When the 0-150 ppm ΔCO_2 category in Model B is split into two categories to produce Model C, the 0-75 ΔCO_2 category has the lowest (fastest) values of log(AHT), after adjusting for the other explanatory variables.

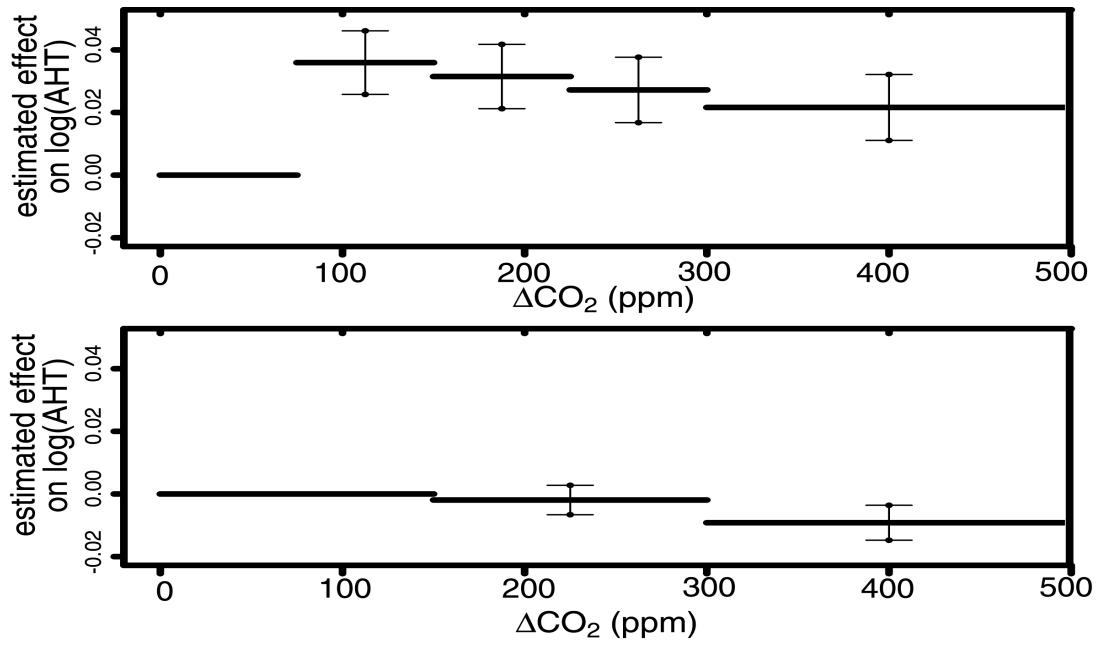

Figure 1. Model coefficients for bins of ΔCO_2 concentration, indicating the effect of ΔCO_2 on log(AHT) with the lowest ΔCO_2 bin used as the reference. The lower and upper plots are results of Model B and Model C, respectively. Horizontal bars indicate ΔCO_2 bin boundaries and vertical error bars represent ± one standard deviation.

We also performed regressions using ventilation rate categories rather than ΔCO_2 categories. There is no evidence for a dependency of AHT on ventilation rate. Even for the highest ventilation rates there is no evidence of reduced AHT. To the extent that there is an apparent ventilation-related effect in this study, it is due to ventilation rate per person (as indicated by ΔCO_2) rather than ventilation per unit indoor volume.

DISCUSSION

We anticipated that performance differences associated with ventilation would be a few percent at most. It is very hard to study causes of such small performance differences in real work. The present study has inadequate statistical power to find effects smaller than about 2%; however, such small changes in productivity could still be economically important.

In the present study, there were only forty half-hour periods (out of 1051) in which ΔCO_2 was below 75 ppm. Nineteen of these periods occurred on a single day (the 78th day of the study, a Friday), and all of the rest occurred during the following week. The entire apparent speed-up in AHT indicated by Model C, for the below-75 ppm bin relative to the other bins, is based on data from only six different days, and 65% of those data are from two consecutive Fridays. Consequently, in spite of the low p-values, we do not consider the results of Model C to

conclusively indicate a faster work-rate when ΔCO_2 was very low and ventilation per worker was very high. If the very high values of ventilation had occurred on 6 days spread throughout the study period, and the same results were found, we would have confidence that the observed effect was really due to ventilation rate. In the present case, though, we are not sure that ventilation is really affecting performance because some uncontrolled and unknown condition, coincident with the periods of very high ventilation rate, could have caused the increased performance.

A limitation of these analyses is the incomplete separation of time of day from ΔCO_2. As time of day increases, ΔCO_2 generally increases (then decreases late in the day); therefore; controlling for time of day via the regression models could have partially obscured a relationship of ΔCO_2 with worker speed. Including "nets" in the regression models may also have partially obscured a relationship of ΔCO_2 with worker speed, because a decrease in work speed caused by higher ΔCO_2 would result in a decrease in "nets".

The results of the present analysis may not apply to other buildings. For instance, it may be that in this building there are no strong indoor sources of pollutants that influence performance, but that in other buildings such sources exist.

CONCLUSIONS

If we exclude periods of very high ventilation rates per occupant (indicated by very low ΔCO_2), we can conclude that the effect of ventilation rate on AHT was less than about 2%. There is some evidence that very high ventilation rates per occupant (very low ΔCO_2) may lead to lower AHT (faster work rates), but the possibility of uncontrolled confounding makes this result less than conclusive in spite of high statistical significance ($p < 0.05$).

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DO INDOOR ENVIRONMENTS IN SCHOOLS INFLUENCE STUDENT PERFORMANCE? A REVIEW OF THE LITERATURE

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ABSTRACT
The goal of this paper was to critically review available evidence on relationships between indoor environmental quality (IEQ) in schools and student performance. Because available evidence from schools was limited, the review expanded to include studies on direct relationships between the performance of children and adults and the indoor environments in schools, workplaces, residences, and controlled laboratory settings. The most persuasive available evidence suggests that some aspects of IEQ, including low ventilation rate and less daylight or light, may reduce the performance of occupants, including students in schools. Other evidence identifies additional possible influences, such as pollen and some carpets. Substantial limitations in the quantity and quality of available research findings suggest many questions for future study. Sufficient evidence is available to justify (1) actions to safeguard IEQ in schools and (2) the conduct of focused, well-designed research to help guide future policies and actions regarding IEQ in schools.

INDEX TERMS: schools, students, performance, indoor environmental quality, indoor air quality

INTRODUCTION
There is widespread concern that indoor environments can affect occupants' health, comfort and performance (U.S. EPA, 2001). Indoor environments in schools are of particular public concern because:

1) Schools, relative to other kinds of buildings, are seen as particularly likely to have environmental deficiencies that could lead to poor indoor environmental quality (IEQ). In particular, chronic shortages of funding in schools contribute to inadequate operation and maintenance of facilities (GAO, 1995).

2) Children breathe higher volumes of air relative to their body weights and are actively growing. Thus, they have greater susceptibility to environmental pollutants than adults. Children also spend more time in school than in any other indoor environment outside the home. Adverse environmental impacts on the learning and performance of students in schools could have important immediate and lifelong effects, for the students and society.

The simple model in Figure 1 shows hypothesized influences of IEQ on the performance of students. Building characteristics [box A] can influence both indoor pollutant exposures and indoor physical parameters (collectively referred to in the paper as IEQ, or as *measured IEQ factors*). Indoor physical parameters can themselves influence indoor pollutant exposures, and both types of IEQ factors can influence health outcomes. Health outcomes can influence performance directly or through effects on attendance. Indoor physical conditions can also directly influence performance.

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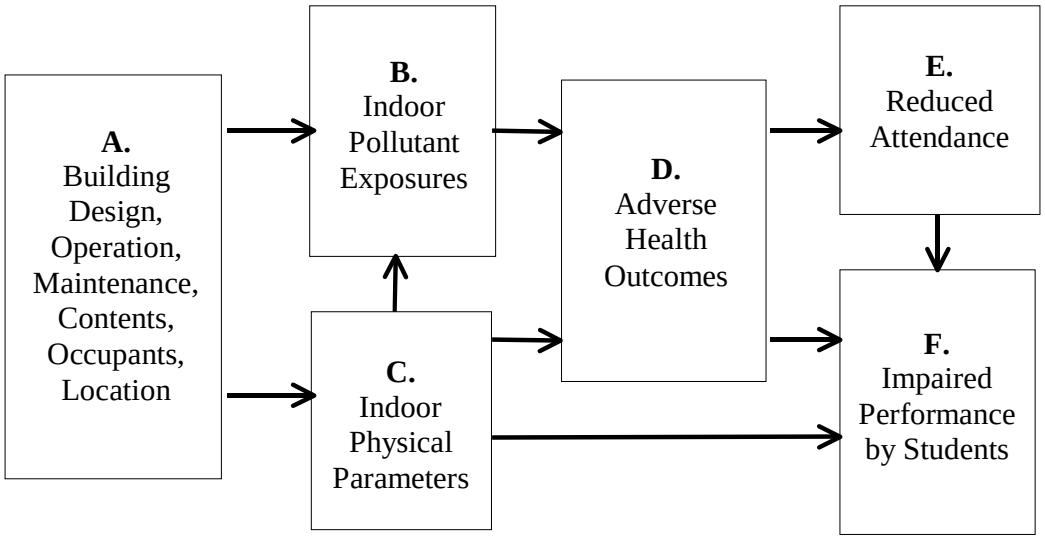

Figure 1. Links in hypothesized causal chains from building characteristics through indoor exposures and physical conditions in schools to student attendance and performance.

This paper summarizes the limited available evidence on direct associations between measured IEQ factors or building characteristics and the performance of building occupants. Although studied as direct associations, these influences are likely to occur through the links shown in Figure 1. Evidence on each of these links contributes to the plausibility of an overall influence of measured IEQ factors or building characteristics upon the performance of building occupants. A larger amount of information is available on these intermediate links; a future report by the same authors will evaluate this evidence.

The present review updates and extends previous reviews on IEQ-performance/productivity links, such as USEPA (2001), Bayer (2000), Fisk (2000), and Sensharma and Woods (1998). Because available evidence from schools was limited, the review included findings on a broader range of subjects and environments relevant to an understanding of IEQ effects on students in schools, e.g., potential adverse effects of school, day-care center, office, and home environments on their occupants. The environmental factors included in this review are: indoor environmental contaminants (including those of outdoor origin and excluding radon, lead, and asbestos); contaminant control processes (e.g., ventilation rate); indoor thermal parameters; and characteristics of buildings that can influence IEQ (e.g., presence of humidification or daylighting).

METHODS

We followed a search strategy similar to Sensharma and Woods (1998), electronically searching (through September 2001) the databases of PubMed, ERIC, Web of Science, Toxline, and the American Society of Heating, Refrigeration and Air-conditioning Engineers (ASHRAE), and manually searching key relevant journals and conference proceedings.

Our review process included evaluation of the strength and consistency of current direct evidence on how IEQ in schools may influence learning or performance of students. We evaluated the strength of studies by evaluating their study designs and measurement methods. Studies with the strongest designs – well-designed experiments, quasi-experiments (e.g., controlled intervention studies), or prospective observational studies, with proper measurements of risks and outcomes and statistical analyses – were considered most

persuasive. Studies with weaker designs or analyses, such as those lacking statistical control for potentially confounding variables, were considered less persuasive. Studies of particularly weak design, such as case studies, uncontrolled interventions, or crude comparisons of two groups, were omitted from the review unless they offered specific information of value. Non-peer-reviewed articles were included despite their usual brevity, lack of available detail, and preliminary nature.

RESULTS

This paper includes information on 21 articles or reports, listed in Tables 1a and 1b. Of these, 14 were peer-reviewed and 6 were considered of strong design. Only one study of the twelve in school settings had a strong design; five studies in other indoor environments had strong designs. This section and Tables 1a and 1b summarize research findings about direct relationships in schools or non-school indoor environments between *measured IEQ factors* and performance (Table 1a) and between *IEQ-related characteristics of buildings* and performance (Table 1b).

Table 1a. Findings from primary research on direct relationships between *measured IEQ factors* and *performance.*

PERFORMANCE OUTCOMES	STUDY FEATURES		MEASURED IEQ FACTORS						Reference
			POLLUTANT EXPOSURES AND CONTROL				PHYSICAL PARAMETERS		
	Setting/Subject	Design	Microbiologic	Chemical	Particles (outdoor source)	Low Ventilation Rate	Higher Temperature	Lower Relative Humidity	
subjective mental performance *	S, C	C-S,©	↓	↓		↓		↓	Smedje 1996
performance tests	L, A	E,©		○					Otto 1992
performance tests	L, A	E,©		○					Molhave 1985
telephone productivity	O, A	PC,©			↓				Burton 2001
reaction and performance tests *	S, C	Q-E,©				↓			Myhrvold 1996
simulated office tasks	O/L, C	E,©				↓			Wargocki 2000
learning efficiency	S/L, C	E,©					↓↑		Pepler 1968
typing	L, C	E,©					↓		Wyon 1974
performance tests	L, C	E,©					↓↑		Wyon 1979

LEGEND for Tables 1a, 1b

Assessed Relationships

- O no statistically significant or noteworthy relationship
- 🡻 statistically significant or noteworthy relationship with adverse outcome
- 🡹 statistically significant or noteworthy relationship with beneficial outcome
- finding from study of strong design

Settings/Subject

- ***S*** ***school***
- **O** office or other non-school workplace
- **L** laboratory
- **C** children (~<18 yrs)
- **A** adults

Performance Outcomes

- * not peer-reviewed

Design

- **E** experiment
- **Q-E** quasi-experiment
- ***PC*** ***prospective cohort***
- **C-S** cross-sectional
- **©** controlled or adjusted for key potential confounder

Table 1b. Findings from primary research on direct relationships between *IEQ-related characteristics of buildings* and *performance.*

PERFORMANCE OUTCOMES	STUDY FEATURES		IEQ-RELATED CHARACTERISTICS OF BUILDINGS										References
			Ventilation Features		Building Features				Interior Features				
	Setting/Subject	Design	Air-Conditioning	Personal Control	Better Facility Condition	Newer Building	Larger Building	Near Noise Source	Carpet	Daylighting	Type of Light Bulb		
achievement tests/ academic progress *	**S, C**	**PC, C-S,©**	🡻	🡹						🡹			Heschong 1999
achievement tests	**S, C**	**Q-E**	🡹										McNall 1967
performance tests	**S/L, C**	**E,©**	🡹										Schoer 1973
achievement tests *	**S, C**	**C-S**	🡹		O🡹								Cash 1993
measured office work	**O, A**	**Q-E,©**		🡹									Kroner 1994
achievement tests *	**S, C**	**C-S, PC**			🡹								Lewis 2000
achievement tests *	**S, C**	**C-S**			🡹								Earthman 1995
achievement tests *	**S, C**	**C-S**			🡹								Berner 1993
subjective mental performance *	**S, C**	**C-S,©**				🡻	🡹						Smedje 1996
reading comprehension	**S, C**	**C-S**						🡻					Haines 2001
simulated office tasks	**O/L, C**	**E,©**							🡻				Wargocki 1999
simulated office tasks*	**O/L, C**	**E,©**							🡻				Lagercrantz 2000
achievement tests	**S, C**	**Q-E**										🡹🡻	Hathaway 1995

Although the available evidence does not persuasively document relationships between performance and specific indoor pollutants, it suggests that lower outdoor air ventilation rates, known to cause generally higher concentrations of the pollutants produced indoors, were related to reduced performance among occupants (Table 1a – Wargocki, 2000; Smedje, 1996). One study documented that an outdoor air pollutant, pollen, can impair indoor performance of sensitized office workers (Table 1a – Burton, 2001). Two studies showed that the presence of a carpet (with uncharacterized emissions) taken from a complaint building impaired performance of occupants (Table 1b – Wargocki, 1999; Lagercrantz 2000). One well-designed study found daylighting, or a related aspect of light in schools, to be related to increased learning by students (Table 1b – Heschong 1999). Insufficient consistent evidence was available to document relationships of indoor thermal parameters or noise to performance.

DISCUSSION

Because many risk factors and outcomes in the area of this review are not well defined, conducting strongly designed research studies has been challenging and the relationships explored here are far from documented causal links. Nevertheless, this review identifies useful scientific findings consistent with IEQ-performance links.

CONCLUSION AND IMPLICATIONS

These findings provide a basis for defining key future research questions and suggest that well-designed research has good prospects for documenting relationships between IEQ and performance in schools. However, since a primary goal of public health research is learning how to prevent adverse effects, effective public health actions do not always require or wait for documented causality; limited scientific evidence combined with common sense and public concern can justify early action. The results of this review may increase the justification for improving IEQ in schools.

ACKNOWLEDGEMENTS

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IMPLEMENTATION OF VOC SOURCE REDUCTION PRACTICES IN A MANUFACTURED HOUSE AND IN SCHOOL CLASSROOMS

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ABSTRACT
Detailed studies of a new manufactured house and four new industrialized relocatable school classrooms were conducted to determine the emission sources of formaldehyde and other VOCs and to identify and implement source reduction practices. Procedures were developed to generate VOC emission factors that allowed reasonably accurate predictions of indoor air VOC concentrations. Based on the identified sources of formaldehyde and other aldehydes, practices were developed to reduce the concentrations of these compounds in new house construction. An alternate ceiling panel reduced formaldehyde concentrations in the classrooms. Overall, the classrooms had relatively low VOC concentrations.

INDEX TERMS
VOCs, Formaldehyde, Source reduction, Manufactured houses, School classrooms

INTRODUCTION
Indoor exposures to toxic volatile organic compounds (VOCs) are of obvious concern. In the U.S., schools have become a focus of complaints regarding children's potential exposures to chemical and biological contaminants including formaldehyde. Odorous VOCs also can adversely affect people's acceptance of indoor environments. In new residences, formaldehyde and odorous aldehydes were among the most prevalent and predominate VOCs (Hodgson et al., 2000). Much less is known about the composition and concentrations of VOCs of concern in new schools. For VOCs with material sources, an efficient way to reduce occupant exposures is to eliminate or modify significant sources. Studies in which we collaborated with a manufactured housing company and a relocatable (RC) classroom manufacturer to implement VOC source identification and reduction practices are reported. The objective of the house study was to identify and verify the major sources of formaldehyde and other VOCs with emphasis on odorous compounds (Hodgson et al., In press). The objective of the school study was to construct two new classrooms using low-emitting interior materials and to compare concentrations of toxic VOCs over time in these classrooms with concentrations in two new standard classrooms serving as controls (Hodgson et al., 2001).

METHODS
Procedures were developed to quantify and assess the emissions of VOCs from interior materials. Specimens of materials of known history were obtained from the building production facilities or from the material manufacturers and then tested for VOC emissions using small-scale chambers following standard guidance (ASTM, 1997). Our method employed specimen conditioning (19 ± 4 days for the house and reduced to 10 days for the schools to improve practicality) prior to a 4-day test with samples collected at the end of

this period to generate VOC emission factors ($\mu g\ m^{-2}\ h^{-1}$) approximating those in a building during early occupancy. Our recommended conditioning and testing methods are summarized in Table 1. Chambers of different sizes may be used; however, the specimen surface area to airflow rate ratio should be kept within a range appropriate for materials covering large areas such as floors and ceilings to establish realistic chamber VOC concentrations.

Table 1. Recommended parameters for testing materials for VOC emissions.

Parameter	Units	Conditioning	Test Period
Temperature	^oC	23 ± 2	23 ± 1
Relative humidity	%RH	50 ± 10	50 ± 5
Chamber volume	m^3	0.01 - 0.08	0.01 - 0.08
Specimen area	m^2	~0.02	~0.02
Area/Flow rate ratio	$m^2 / m^3\ h^{-1}$	0.25 - 0.45	0.25 - 0.45
Duration	h	≥240	96

Since previously we had identified the predominant VOCs in new manufactured houses from the same facility in Florida (Hodgson et al., 2000), the house study focused on wood and wood products, which were the most likely sources of formaldehyde and odorous aldehydes. These materials included cabinetry components, interior doors and the plywood subfloor. Emission rates of toxic and odorous VOCs attributable to each major material to be used in the test house were calculated from the measured emission factors and the quantity of material installed in the house. These emission rates were summed by compound to predict whole-house emission rates. Indoor and outdoor aldehyde and VOC measurements were made three months after the house was installed at a sales center. The house ventilation rate was at steady-state conditions and was quantified concurrently by SF_6 tracer gas decay. Whole-house emission rates were derived by steady-state mass balance from the concentrations and the house ventilation rate and volume.

Table 2. Toxic chemicals of concern in California.

California Regulatory Lists of Toxic Chemicals	www.URL
Air Toxics Hot Spots Program Risk Assessment Guidelines; Chemicals with Established Noncancer Chronic Reference Exposure Levels (RELs)	oehha.org/air/ chronic_rels/
Safe Drinking Water and Toxic Enforcement Act of 1986 (Proposition 65); Chemicals Known to the State to Cause Cancer or Reproductive Toxicity	oehha.org/prop 65/prop65_list/
Substances Identified as Toxic Air Contaminants (TACs) by the Air Resources Board (includes all U.S. EPA Hazardous Air Pollutants)	arb.ca.gov/ toxics/taclist.htm

Working with a California manufacturer of RC classrooms, a study was conducted to measure VOC emissions from a number of standard and alternate interior finish materials (Hodgson et al., 2001). Alternate carpets, wall panels and ceiling panels were selected for use in the construction of two modified classrooms based on these results and considerations of cost, performance and maintenance. Emphasis was placed on reducing indoor concentrations of VOCs that appear on California regulatory lists of toxic chemicals (Table 2). VOC concentrations were estimated for these classrooms and two standard classrooms based on the measured emission factors, material quantities, building

volumes, and design ventilation rates based on expected occupancy. The four classrooms subsequently were constructed for a field study. For a case-crossover design, each classroom was equipped with a standard HVAC system and an advanced system employing indirect/direct evaporative cooling (IDEC) to provide continuous ventilation of at least 7 L s^{-1} per occupant (Apte et al., 2001). At each of two schools, a modified classroom was sited adjacent to a standard classroom. In the fall semester, integrated school-day indoor and outdoor aldehyde and VOC measurements were made weekly for eight weeks with HVAC systems switched on alternate weeks. Energy and environmental parameters were measured continuously. At the beginning and end of the 8-week period, classroom DEC systems were operated at steady-state conditions during unoccupied hours. Indoor and outdoor aldehyde and VOC concentrations were measured and the ventilation rates were quantified by CO_2 tracer decay. Whole-building emission rates were derived from these data by mass balance.

RESULTS AND DISCUSSION

A large mass and bare (unfinished) surface area of composite wood products were used in the production of the house kitchen, utility and bathroom cabinets (Table 3). The top surfaces of the countertops were finished with laminate. The other composite materials had decorative polyvinyl chloride (PVC) applied to one or more surfaces. The other predominant wood products in the house were the passage doors (molded high-density fiberboard with acrylic finish) and the plywood subfloor under carpeted areas (Table 3). Bare particleboard (PB) and medium density fiberboard (MDF) surfaces had relatively high formaldehyde emission factors. The finished surfaces (laminate and PVC) of these materials had substantially lower emission factors. The installation of a standard carpet and cushion over the plywood had no significant effect on the emission factors of most aldehydes including formaldehyde. The cabinetry materials and passage doors were the largest predicted contributors to formaldehyde whole-house emission rates; the subfloor was the largest predicted source of hexanal, an odorous aldehyde (Table 3).

Table 3. Quantities of composite wood products used in new house and predicted whole-house emission rates of formaldehyde and hexanal from these sources.

Material Description	Mass (kg)	Bare Area (m^2)	Emission Rate (mg h^{-1}) Formald.	Hexanal
PB[a] counter top, underside	155	12.1	1.0	2.7
PB case, 1 side	74	9.2	4.3	--
MDF[b] stile, 1 of 4 sides	72	4.9	1.6	1.2
Hardboard, 1 side	39	11.7	0.1	--
Cabinetry Totals	340	38	7.0	3.9
Passage doors	--	25	3.8	1.1
Plywood subfloor[c]	1,270	111	1.1	18.3

[a]PB = particleboard; [b]MDF = medium-density fiberboard; [c]Area overlain by carpet.

Concentrations of selected VOCs measured in the house at a ventilation rate of 0.28 h^{-1} three months after installation are presented in Table 4 along with a comparison of derived and predicted whole-house emission rates. The formaldehyde concentration was within the range of typical maximum guideline values of 50-100 ppb. The concentrations of many

higher molecular weight aldehydes exceeded their odor thresholds (Devos et al, 1990). For 10 of the 14 compounds including formaldehyde, the average predicted rates were within a factor of ±2 of the derived rates, suggesting that many of the sources of these compounds were correctly accounted for.

Concentrations of formaldehyde and odorous aldehydes can be reduced in new manufactured house construction by several simple practices directed at cabinet construction and other sources (Table 5). A PVC-coated door of identical design was found to reduce formaldehyde emissions from this source to a negligible level (<0.1 mg h^{-1}). Overlaying plywood with a carpet cushion with an integral spill barrier or a sheet barrier material was partially effective for reducing aldehyde emissions. However, the potential for such materials to create moisture-related problems in hot-humid climates requires investigation.

Table 4. Concentrations and emission rates of terpenes and aldehydes in new house.

Compound	In-Out Conc (ppb)	Odor Threshold[a] (ppb)	Derived ER[b] (mg h^{-1})	Predicted ER[c] (mg h^{-1})	Pred/Deriv w/in 2 x? Y/N[d]
α-Pinene	42	690	25 ± 4	12.9-31	Y
β-Pinene	13.3	--	8.1 ± 1.3	3.4-7.8	Y
d-Limonene	7.2	44	4.4 ± 0.7	3.3-12.6	Y
Formaldehyde	74	870	9.9 ±1.6	11.7-12.2	Y
Acetaldehyde	21	186	4.2 ±0.7	2.0-3.0	Y
Pentanal	21	6.0	8.0 ± 1.3	3.7-4.1	N
Hexanal	65	13.8	29 ± 5	22-24	Y
2-Furaldehyde	3.9	780	1.69 ± 0.27	0.94	Y
Heptanal	5.3	4.8	2.7 ± 0.4	0.53-0.59	N
2-Heptenal	3.0	13.5/6.0[e]	1.51 ± 0.24	0.61-0.69	N
Benzaldehyde	3.1	42	1.46 ± 0.23	1.06	Y
Octanal	8.2	1.3	4.7 ± 0.7	1.19-1.29	N
2-Octenal	3.7	2.0/0.7[e]	2.1 ± 0.3	1.52-1.69	Y
Nonanal	7.0	2.2	4.5 ± 0.7	2.8-3.0	Y

[a]Devos et al., 1990; [b]Emission rates (ERs) ± 1 stdev. derived from concentrations and house parameters; [c]Predicted whole-house ERs are sums of ERs for wood product sources; [d]Y/N = Yes/No, predicted ER is within factor of two of calculated ER; [e]*cis* isomers of 2-heptanal and
2-octenal have lower odor thresholds than *trans* isomers.

Table 5. Recommended VOC source reduction practices for new house construction.

No.	Source Reduction Practice
1	When alternates exist, avoid wood products with urea-formaldehyde resin system
2	Construct cabinet cases with fully encapsulated wood products
3	Use frameless cabinets to eliminate MDF stiles
4	Apply laminate backing sheet to undersides of PB countertops
5	Use alternate low-formaldehyde emitting passage doors
6	Apply barrier material over plywood subfloor in carpeted areas (see caution in text)

The laboratory study of RC classroom materials showed that the standard and alternate carpets generally were not significant sources of VOCs of concern. Also, glue-down carpet

systems reduced the emissions of formaldehyde from the subfloor. Fiberglass ceiling panels were identified as the major formaldehyde source among the standard materials. The emission factor was 32 $\mu g\ m^{-2}\ h^{-1}$. Two mineral-fiber ceiling panels were tested. One had emissions comparable to the fiberglass material, while the other had no detectable formaldehyde emissions. The classroom walls were entirely covered with tackable panels. Five panel systems were tested. The standard wood fiber panel covered with PVC fabric emitted acetaldehyde, phenol, 2-(2-butoxyethoxy)ethanol, vinyl acetate, 1,24-trimethylbenzene and
1-methyl-2-pyrrolidinone. The selected alternate, which was identical except that the PVC fabric was Teflon coated, was predicted to reduce concentrations of these compounds, while increasing toluene concentrations by ~1 ppb during occupancy.
RC formaldehyde concentrations were higher than predicted by the laboratory study. The formaldehyde emission rates in the four classrooms derived from measurements made at the beginning (pre) and end (post) of the 8-week study are shown in Table 6. At the end, there was an aberrantly high emission rate in RC 4. This was attributed to an as yet to be identified source added to the room during the study. Ignoring this value, the formaldehyde emission rates decreased over the course of the study, and the modified classrooms had lower emission rates than their paired standard control classrooms. Concentrations were predicted for the classrooms based on the average measured IDEC airflow rate for each school. These were higher for School A due to higher occupancy. The predicted values are generally consistent with the measured concentration ranges. The higher concentrations in control classrooms RC 1c and RC 3c occurred during days when their IDEC systems were not operated continuously.

Table 6. Derived formaldehyde emission rates and predicted formaldehyde concentrations in four new classrooms operating with IDEC system. Emission studies were conducted prior to (pre) and after (post) 8 weeks of occupancy. Predictions were made using respective average airflow rates of 850 and 675 $m^3\ h^{-1}$ for Schools A and B and are compared to concentrations measured over four weeks of classroom use with IDEC system. Rooms RC 2 and RC 4 had ceiling panels with negligible formaldehyde emissions. Rooms RC 1c and RC 3c are controls.

	Derived Emission rate ($\mu g\ h^{-1}$)		Indoor-Outdoor Concentration (ppb) Predicted		Measured during use	
School, Room	Pre	Post	Pre	Post	Average	Range
School A, RC 1c,	9,900	3,450	9.5	3.3	9.9	3.5 – 19.1
School A, RC 2	7,040	2,660	6.7	2.5	4.1	3.0 – 4.7
School B, RC 3c	3,800	1,310	4.6	1.6	6.1	2.8 – 10.6
School B, RC 4	2,370	26,800*	2.9	32.3	12.1	8.4 – 17.4

*Attributed to unidentified source added after fall semester began.

Table 7 compares the concentrations of toxic VOCs other than formaldehyde in the four classrooms over the four weeks in which the IDEC system was used. Some additional toxic VOCs (not shown) were measured at generally lower concentrations. The concentration ranges for paired modified and control classrooms overlap, with several exceptions. 1-Methyl-2-pyrrolidinone and phenol (School B only) concentrations were lower in the modified classrooms probably due to the use of the alternate wall panel. Caprolactam was highest in one control classroom. The likely source was the Nylon 6

fiber carpet installed in that room, but not included in the laboratory study. Toluene differences were small as predicted.

Table 7. Concentrations of other toxic VOCs in four new classrooms over four weeks of use with IDEC system. Rooms RC 2 and RC 4 are modified; RC 1c and RC 3c are controls.

	Indoor-Outdoor Concentration (ppb)				
	Study	School A		School B	
Compound	Median	RC 1c	RC 2	RC 3c	RC 4
Toluene	0.7	0.2-0.6	0.3-0.7	0.3-1.0	0.5-1.9
2-Propanol	2.6	2.1-6.6	2.4-6.7	2.0-5.0	2.0-5.6
Phenol	1.0	0.8-2.5	0.3-0.8	0.2-1.4	<0.1-1.0
Vinyl acetate	0.1	<0.1-0.2	<0.1	<0.1	0.1-0.4
Acetaldehyde	3.1	0.1-4.7	0.2-1.3	<0.1-2.9	0.4-5.2
2-Butanone	0.3	0.1-0.4	0.1-0.3	<0.1-0.4	0.1-0.4
1-Methyl-2-pyrrolidinone	0.2	0.2-0.5	<0.1	0.2-1.8	<0.1-0.1
Caprolactam	0.2	0.1-0.2	<0.1-0.1	2.3-6.5	0.1-0.3

CONCLUSIONS

The composition and concentrations of VOCs in the manufactured house and the classrooms were substantially different. The house had relatively elevated concentrations of formaldehyde and acetaldehyde and of odorous aldehydes that often exceeded odor thresholds. The school study focused on listed toxic chemicals, but also included the analysis of odorous aldehydes. The concentrations of these compounds were low. The differences between the house and the classrooms largely were due to the much higher ventilation rates and the use of higher quality, fully encapsulated cabinetry and generally less wood products in the classrooms. Although the school data have yet to be fully analyzed, several small improvements in the concentrations of toxic VOCs can probably be attributed to the use of alternate materials. The use of the alternate wall panel had a small impact on the concentrations of several VOCs. The use of a lower emitting ceiling panel probably reduced formaldehyde concentrations.

These studies found that the systematic identification of VOC sources and the implementation of source reduction practices can be effectively implemented in industrialized buildings. As the process was shown to be relatively accurate, manufacturers can be assured that if they test the standard and new materials under consideration, most problematic sources of VOC contamination will be identified and eliminated. Because manufacturers construct large numbers of units, which often incorporate the same interior materials, the costs are not likely to be prohibitive due to economies of scale.

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BUILDING-RELATED RISK FACTORS AND WORK-RELATED LOWER RESPIRATORY SYMPTOMS IN 80 OFFICE BUILDINGS

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ABSTRACT

We assessed building-related risk factors for lower respiratory symptoms in office workers. The National Institute for Occupational Safety and Health in 1993 collected data during indoor environmental health investigations of workplaces. We used multivariate logistic regression analyses to assess relationships between lower respiratory symptoms in office workers and risk factors plausibly related to microbiologic contamination. Among 2,435 occupants in 80 office buildings, frequent, work-related multiple lower respiratory symptoms were strongly associated, in multivariate models, with two risk factors for microbiologic contamination: poor pan drainage under cooling coils and debris in outside air intake. Associations tended to be stronger among those with a history of physician-diagnosed asthma. These findings suggest that adverse lower respiratory health effects from indoor work environments, although unusual, may occur in relation to poorly designed or maintained ventilation systems, particularly among previously diagnosed asthmatics. These findings require confirmation in more representative buildings.

INDEX TERMS

Indoor air quality, ventilation systems, symptoms, respiratory health, asthma

INTRODUCTION

Documented building-related respiratory disease such as hypersensitivity pneumonitis or humidifier fever associated with microbiologic contamination of the indoor environment has been reported occasionally in indoor, nonindustrial workplaces such as office buildings (e.g., Kreiss, 1989; Hodgson *et al*., 1987; Seuri *et al.*, 2000). In contrast, episodes of nonspecific health complaints in indoor workplaces (sometimes called sick building syndrome or nonspecific building-related symptoms), not attributable to specific recognized disease or exposures, have been commonly reported in recent decades (Mendell, 1993). Although specific causal exposures for nonspecific building-related symptoms have not yet been established, research has identified a number of person-, job-, workplace-, and building-related risk factors for these symptoms (e.g., air-conditioning systems, low ventilation rate, high temperature, dust, endotoxin) (Mendell, 1993; Gyntelberg *et al.*, 1994; Teeuw *et al.*, 1994; Bornehag *et al.*, 2001). Few of these reports have considered lower respiratory symptoms, the least common symptoms reported in indoor work environments (Mendell, 1993; Mendell *et al.*,1996; Malkin *et al.*, 1996). Some recent studies have associated risk factors in non-industrial indoor work environments with increased work-related lower respiratory symptoms (Mendell *et al*.,1996a; Sieber *et al.*, 1996; Ruotsalainen *et al.*, 1995).

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We explored relationships within existing data on risk factors and symptoms in office buildings. For this analysis, we hypothesized that microbiologic contamination of indoor spaces or ventilation systems in office buildings may cause or exacerbate *unrecognized building-related respiratory disease* that presents as *work-related lower respiratory symptoms*. We also hypothesized that associations between these symptom outcomes and risk factors would be stronger among doctor-diagnosed asthmatics, due to increased sensitivity among prior asthmatics and among any with new-onset asthma caused by exposures at work.

METHODS

Data Collection

Methods used in conducting the surveys (Crandall and Sieber, 1996) are briefly discussed below. Between April and July 1993, NIOSH investigators collected standardized health, building, and environmental data from a randomly chosen evaluation area within each of 105 U.S. office buildings. Building were selected from over 800 buildings that requested health hazard investigations from NIOSH. A self-administered questionnaire was used to assess health histories and symptoms in workers. Industrial hygienists assessed characteristics of the study buildings, including their ventilation systems and their indoor environments, using a standardized inspection form and simple measurements. Complete data collected according to the study protocol were available from 80 office buildings and 2345 workers.

Outcome and risk variables in analyses

The full analysis included twenty outcome definitions constructed from four lower respiratory symptoms – wheeze, shortness of breath, chest tightness, and cough. Definitions required symptom occurrence at least once per week in the previous four weeks and improvement away from the workplace, as in Sieber *et al.* (1996). We report here only results for the outcome definition used in previous analyses by Sieber *et al.* (1996), there called "frequent multiple lower respiratory symptoms," but here called "at least three of four frequent, work-related (FWR) lower respiratory symptoms."

Twenty dichotomous building-related risk factors were selected for these analyses (Table 1): those previously associated with work-related lower respiratory symptoms in partially adjusted regression models (Sieber *et al.*, 1996), hypothetically related to risk of microbiologic contamination, and having adequate data for analysis. Regression models also included dichotomous personal risk factor variables for gender, age, smoking, and asthma. A set of similar models with interaction terms assessed whether history of asthma influenced the relationships of risk factors to outcomes.

Preliminary models

We used multivariate logistic regression models to estimate adjusted odds ratios (ORs), a measure of strength of association for which ORs >1.0 imply increased risk. To decrease the large number of inter-correlated independent risk variables, we first screened each in a regression model with only personal covariates for gender, age, smoking, and asthma status. We included in further analyses only the risk variables with p-values <0.20 in screening models. We then created preliminary models for four subgroups of risk variables: potential sources of microbiological contaminants less than 25 feet from the outside air intake;problems

with particle filtration in the heating, ventilation, and air-conditioning (HVAC) system; dirt in the HVAC system; and moisture in the HVAC system (see Table 1). Three variables - no scheduled HVAC inspection, water damage in the workspace, and indoor surface dusting daily -- were considered by themselves without inclusion in a sub-group. Each subgroup model also included personal covariates for gender, age, smoking, and asthma status. From each subgroup, variables for which p>0.20 were eliminated sequentially. A final model was created containing the selected risk factor variables from the reduced subgroup models, the ungrouped risk factor variables, and gender, age, smoking, and asthma variables. This full model was reduced by elimination of terms for which p>0.10.

We added interaction variables for (building risk factor * asthma history) to selected partially adjusted models and to final reduced models.

RESULTS

Among models constructed with terms for interaction between asthma history and building risk factors, only the partially adjusted models containing single risk factors converged. Most ORs among asthmatics were higher than among non-asthmatics (Figure 1), although p-values exceeded 0.05 for all the interaction terms.

For single environmental risk factors included in partially

Table 1. Building-related risk factors assessed in initial multivariate models

Outdoor Air Intake Near Contaminant Sources:
- Standing water [a]
- Exhaust vents [a]
- Sanitary vents [a]
- Trash dumpster

Dirt in HVAC
- Dusty air handling housing
- Dirty sound liner
- Debris inside air intake [a]
- Coils dirty
- Residue/dirt in drain pans
- Dirty duct work
- Dirty duct liner

Air Filter Problem
- Filter not secure in place [a]
- Dirty filters [a]
- Limited or no access for changing or inspection [a]
- Poor filter fit in frames

Moisture in HVAC
- Moist sound liner
- Poor or no drainage from pans [a]

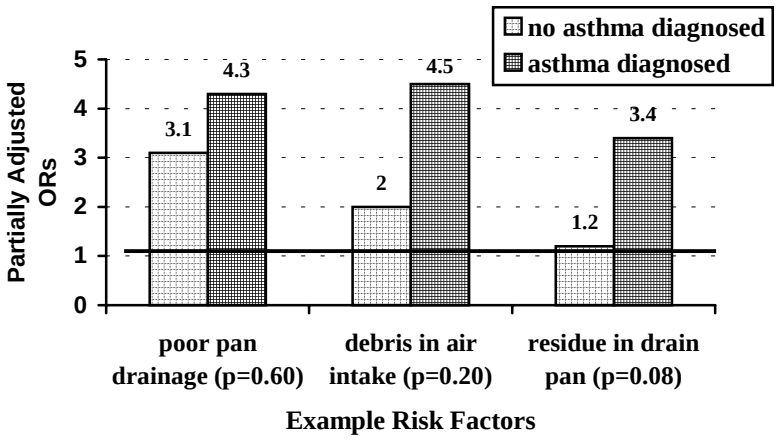
No scheduled HVAC inspection [a]

Workspace Moisture Incursion
- Water damage in workspace [a]

Workspace Maintenance
- Surface dusting daily [a]

[a] p-value<0.20 for association with both outcomes – at least 3 of 4 FWR respiratory symptoms; FWR wheeze, shortness of breath, and cough – in partially adjusted models containing this subgroup of risk factors

Figure 1. History of asthma as effect modifier for risk of at least 3 of 4 FWR lower respiratory symptoms in office buildings studied by NIOSH



adjusted models, lower confidence limits for 10 of the 20 factors exceeded 1.0, and for the two potentially protective factors upper confidence limits equaled 1.0. Table 2 shows estimates for variables in the final model, including two building-related risk factors: debris in air intake and poor pan drainage.

Table2. Odds ratios (OR) and 95% confidence intervals (CI) for building risk factors from final multivariate models

	At least 3 of 4 FWR symptoms		**FWR wheeze, shortness of breath, *and* cough**	
Independent Variable	OR	(95% CI)	OR	(95% CI)
Dirt in HVAC variables:				
Debris inside air intake	2.0	(1.0-3.9)	3.6	(1.4-9.4)
Moisture in HVAC variables:				
Poor pan drainage	2.6	(1.3-5.2)	2.8	(1.1-7.4)
Personal variables:				
female gender	3.0	(1.4-6.6)	3.2	(1.0-9.8)
age over 40 years	2.3	(1.2-4.5)	2.2	(0.8-5.9)
ever smoked	1.2	(0.6-2.1)	1.8	(0.7-4.2)
dr.-diagnosed asthma	8.0	(4.4-14)	6.9	(3.0-16)

DISCUSSION

The multivariate-adjusted analysis found strong associations between frequent, work-related multiple lower respiratory symptoms in office workers and two building-related risk factors: poorly draining drain pans under cooling coils and debris inside outdoor air intake (Table 2). This finding extends previously reported findings by Sieber *et al.* (1996) from these same data. Sieber *et al.* reported relative risks for associations between three kinds of respiratory health outcomes, including the one assessed here, and a number of risk factors in office buildings. Sieber *et al.* adjusted the estimates for each risk factor, however, only for age and gender, not for other personal factors or for any building-related risk factors. Analyses here adjusted simultaneously for personal factors (Table 2) and other building-related risk factors.

The two building-related factors included in the final models – "poor pan drainage" and "debris inside air intake" – indicate conditions that would allow growth of microbiological organisms and dissemination of their products – allergens, irritants, or toxins -- through the ventilation system. Because of correlations between the 20 risk factors (discussed in Sieber *et al.* (1996)), replication of these analyses in larger data sets is necessary to better characterize the environmental risks.

The tendency towards increased susceptibility to microbiologic risk factors (of which subjects were not aware) among previously diagnosed asthmatics (Figure 1) is noteworthy. For instance, for "residue in drain pan," the ORs of 3.4 among asthmatics vs. 1.2 among non-asthmatics for the outcome "at least three or four FWR lower respiratory symptoms" represents a much greater increase in risk among asthmatics (p=0.08 for this difference). Although differences were less dramatic (Figure 1) for the two risk factors included in the final multivariate models, reassessment of this finding in larger populations is essential.

Limitations of study

The office buildings included in this analysis, all buildings for which health hazard investigations had been requested from NIOSH, may differ from other buildings. Findings thus should not be extrapolated to the general population of office buildings unless replicated in more general studies. These "complaint" buildings nevertheless represent an important set of buildings requiring resolution of environmentally related health complaints. Occupants' concerns about effects of their indoor environments may have exaggerated estimates of symptom prevalence in these buildings. However, these concerns could not create apparent associations of symptoms with risk factors of which respondents were unaware, such as conditions within the ventilation systems.

CONCLUSIONS AND IMPLICATIONS

Although work-related lower respiratory symptoms reported in industrial or agricultural settings are considered potential indicators of work-related disease, such symptoms in non-industrial indoor environments are generally accorded little clinical significance because of the presumed lack of causal exposures. Yet multiple reports have described serious respiratory disease, caused by building or HVAC- related moisture and mold in workplaces (Kreiss, 1989; Woodard *et al.*, 1988; Seuri *et al.*, 2000; Hoffman *et al.*, 1993; Thörn, 1996; Jarvis and Morey, 2001).

The findings here, although requiring replication, suggest that building-related respiratory health effects may occur without recognition among a subset of indoor workers with moisture- or contaminant-related exposures. A history of asthma may confer heightened susceptibility to these exposures, or asthma *caused* by exposures in the building may be chronically exacerbated by continuing exposure. Even if the proportions of buildings and of workers with such exposures were small, the absolute numbers, among the almost 70 million US workers in indoor environments, would be of public health significance.

Replication of the findings reported here would provide an initial method for identifying indoor workers at increased risk for work-related respiratory health effects. Investigators of health complaints in buildings often assess and describe only the most commonly reported symptoms such as headache, fatigue, or eye irritation, excluding the rarer lower respiratory symptoms. Yet this exclusion, as suggested in the findings here, may hide an important biologic response in a key susceptible subpopulation. The symptoms studied here may represent unrecognized sub-clinical manifestations of known building-related disease such as asthma or hypersensitivity pneumonitis (Kreiss, 1989).

Our findings show that moisture-and contaminant-related risks in indoor work environments are associated with work-related respiratory health effects among the workers. History of asthma may confer increased susceptibility to, or indicate initial sensitization by, microbiologic exposures associated with risk factors such as poor pan drainage and debris in air intake. Future research, possibly focused within susceptible subgroups, on occupants with multiple lower respiratory symptoms exacerbated within buildings may help identify more specific risks for building-related respiratory disease.

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V. FACTORS AFFECTING INDOOR EXPOSURE TO PARTICLES OF OUTDOOR ORIGIN

PREDICTING INDOOR PM2.5 OF OUTDOOR ORIGIN: TESTING A TRANSIENT SIZE-RESOLVED MODEL USING INTENSIVE MEASUREMENTS FROM A RESIDENCE

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ABSTRACT
We report tests of a model for indoor PM2.5 of outdoor origin that incorporates physical mechanisms for time dependent transport, and size dependent penetration and deposition. This work was performed using information obtained from an intensive study of a house near Fresno, CA, USA. During the multi-week study covering two seasons, we measured particles in both indoor and outdoor air, with high temporal, chemical, and size resolution, and other variables that also affect transport and fate. Results suggest that 1) the model captures a significant fraction of the variation in meteorologically forced air infiltration rate, 2) the predicted indoor/outdoor PM2.5 ratio is not consistent with the measured ratio unless a large (unphysical) deposition rate > 2 hr^{-1} is assumed, and 3) the differences between model and measurement in indoor PM2.5 are likely due to loss of volatile ammonium-nitrate aerosol. We conclude that nitrate particle volitization must be included in the model formulation.

INDEX TERMS
Measurements, Models, Outdoor, Particulate Matter, Ventilation

INTRODUCTION
Exposure to air containing particulate matter smaller than 2.5 microns in size (PM2.5) has been implicated in increased mortality and morbidity (Samet et al., 2000). Because personal exposures to outdoor sources of PM2.5 are better correlated with indoor than outdoor concentrationn, models have been developed to estimate indoor concentrations of outdoor origin (Thornburg et al., 2001; Riley et al., 2002). In particular, recent work by Riley et al. (2002, hereafter RMLN) incorporated current understanding of physical factors governing the indoor proportion of outdoor particles (IPOP), and the typical size distributions of outdoor particles, to estimate steady state probability distributions for IPOP and PM2.5 for urban and rural residences and small commercial buildings. This work showed that typically $0.3 < IPOP < 1$, which is consistent with earlier work. Herein, we construct and test a transient physical model based on parameterizations from RMLN, and compare the results with measurements made at a residential research house.

METHODS

The building used for this study is an unoccupied single story (134 m^2 area) suburban residence in Clovis (a suburb of Fresno), CA. Measurements were made from August, 2000 to January, 2001, with three intensive measurement periods in October and December, 2000, and January, 2001. More information on the instrumentation and experimental conditions is provided in Lunden, et al. (2002a,b) and Thatcher et al. (2002a).

The model used to estimate indoor PM2.5 of outdoor origin is expressed as a 1st order time dependent linear differential equation for the concentration of indoor PM, $C_{in,j}$, in terms of outdoor concentration, $C_{out,j}$,

$$dC_{in,j}/dt = P_j(\Delta p,x)\, C_{out,j}\, \lambda - C_{in}\,(\beta_j(x) + \lambda), \text{ where} \qquad (1)$$

the air infiltration rate into the building λ, the fraction of particles penetrating the building shell $P_j(\Delta p,x)$, and the particle deposition rate inside the building $\beta_j(x)$ are functions of particle size j, indoor/outdoor pressure difference, Δp, and other building factors, x, such as building furnishings and the turbulence intensity of indoor air. Particle penetration factor and deposition rate were estimated using the work of Liu and Nazaroff (2000), and Lai and Nazaroff (2001) and Thatcher et al. (2002b) respectively, under assumptions similar to those in RMLN. Curves showing typical ranges for size dependent penetration and deposition rate are shown in Figure 1. Taken together, these curves show that 0.1 to ~ 1 μm particles are expected to transport into a building and deposit at rates in a range (0.1 – 1 hr^{-1}) that is comparable to typical residential ventilation rates.

The air infiltration rate into the house is predicted for periods without active mechanical forcing using the LBNL/AIM infiltration model (Walker and Wilson, 1998), measured indoor-outdoor temperature differences, roof level wind speed and direction, and building leakage area as measured with the blower door method (Dickerhoff, 2001). The predicted infiltration rate is compared with the measured infiltration rate determined with a continuous SF_6 tracer gas measurement.

Indoor and outdoor particle number data were measured every 5 minutes with matched light scattering and aerodynamic particle sizing instruments, and binned into 20 logarithmically spaced bins from 0.1 to 2.5μm in size (Lunden, 2002b). Predicted indoor particle numbers are then calculated using the measured outdoor particle number data, the size dependent parameters from Figure 1, and the measured and predicted infiltration rates from Figure 2 as inputs to a forward marching finite difference form of Eq (1). Total PM2.5 concentration is calculated as the sum of size resolved particulate numbers assuming spherical particles with uniform density. Note that although uncertainty in the estimate of mass concentration is expected (due to uncertainties in the absolute instrument collection efficiency, particle shape and density), the estimated indoor/outdoor PM2.5 ratio is somewhat insensitive to these errors. Finally, concentrations are averaged over 12 hr intervals.

RESULTS

Measured and predicted ventilation rates from a period in December, 2000 to January, 2001, are shown in Figure 2. After active experimental manipulations of the house in late Dec., the air infiltration was dominated by thermal and wind induced indoor/outdoor pressure differentials (see inset of Figure 2). Linear regression of predicted on measured infiltration rates for this later period reveals that the predicted infiltration explains 64% of the variance in the measured data with a best fit slope of 0.65 +/- 0.02 and negligible offset. This suggests that the infiltration model captures most of the meteorologically driven variations but that a multiplicative uncertainty is present in some combination of the estimates of the model coefficients, building parameters, or measured air exchange rate. Below, we examine the effect of uncertainty in infiltration rate on the predicted indoor/outdoor PM2.5 ratio.

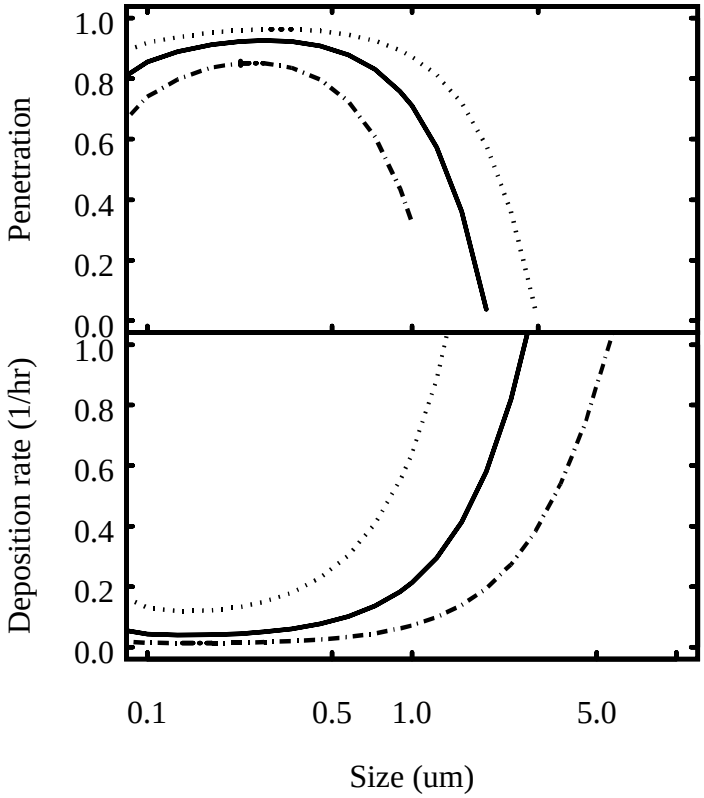


Figure 1. Size dependent particle penetration and depositions rates. Penetration is shown for indoor/outdoor pressures differences of 1 Pa (dotted), 0.3 Pa (solid), and 0.1 Pa (dashed). Deposition rate is shown as a mean estimate (solid) within a factor of 3 variations indicative of range of estimates from literature.

The measured and predicted ratios of indoor to outdoor PM2.5 are shown in Figure 3. In contrast to expectations, the values were measured to be significantly less than unity except during periods when outdoor air was rapidly forced into the building. The low ratio can, however, be explained if a very high ($> 2\ hr^{-1}$) deposition rate is adopted. Additionally, the lower panel of Figure 3 shows that the uncertainty introduced by the predicted versus measured infiltration rate is relatively small. Similarly, using the upper estimate of deposition rates from Figure 1 does not have a very strong influence on the

indoor/outdoor ratio. These two results can be explained by examining the steady state solution to Eq (1), $C_{in,j}$ (steady state) = $C_{out,j}$ λ /(λ+β). When infiltration and deposition are approximately equal variations in either parameter have a less than linear effect on the indoor/outdoor ratio.

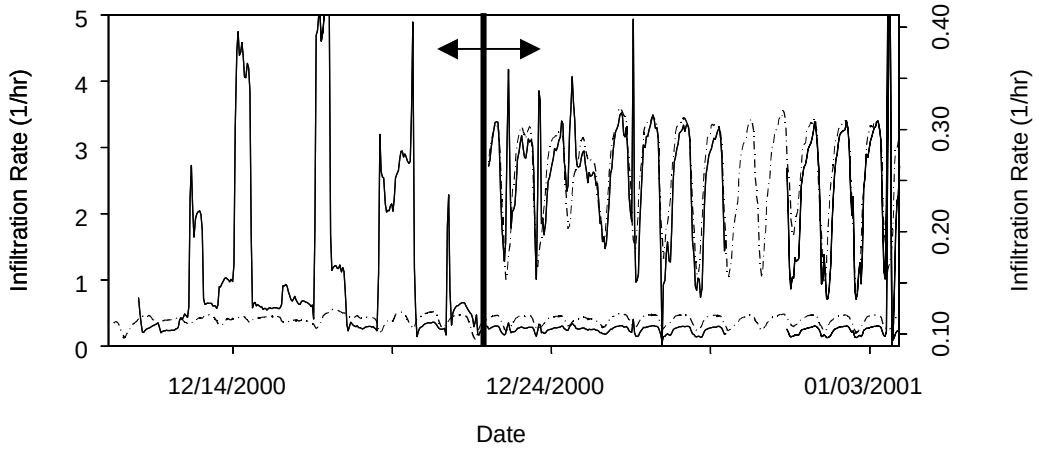


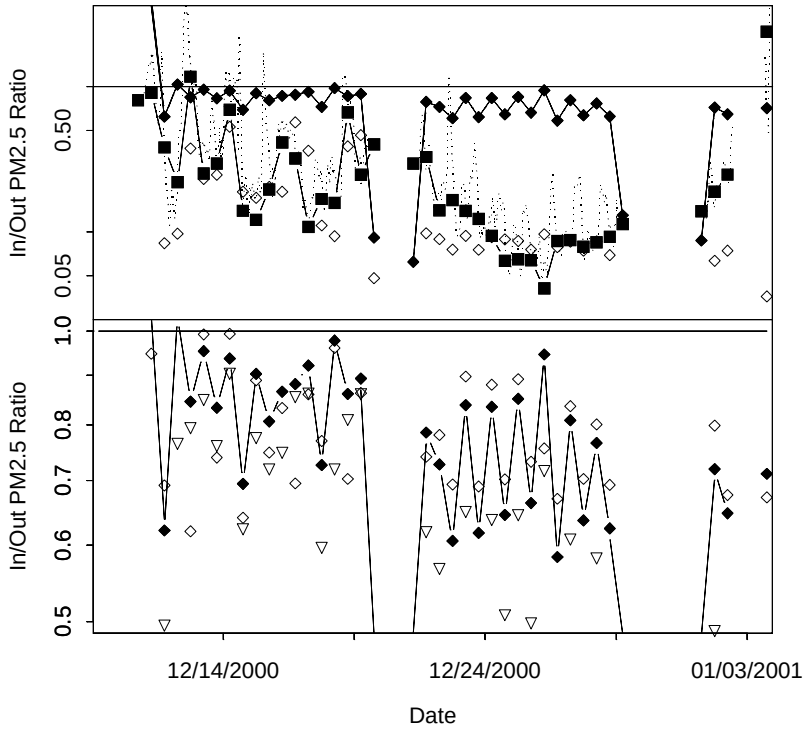
Figure 2. Measured (solid line) and predicted (dashed line) air exchange rates for period of winter measurements. To show the correlation between measured and predicted infiltration rate when building was not mechanically forced, the right portion of the graph is magnified (see right hand scale) and shows the measured and predicted infiltration rates with the predicted values scaled by a factor of 0.65.

DISCUSSION

The principal result of this work is that the measured indoor/outdoor PM2.5 ratio was significantly lower than can be explained by current information on the transport and loss of non-volatile aerosol species. This is likely due to the prevalence of volatile ammonium nitrate aerosol that vaporizes upon entry into the house (Lunden et al. (2002a)). The results obtained here are also consistent with those of Thatcher et al. (2002a), where indoor/outdoor differences in sulfate aerosols are well predicted using the physically based model of Eq (1) to obtain best fit penetration and deposition rates in the range given in Figure 1, while a high deposition rate was necessary to fit the measured indoor nitrate aerosol. Where volatile species constitute a significant fraction of total fine aerosol, successful predictions of indoor PM2.5 require a model that includes gas-particle partitioning.

These results also suggest that natural infiltration rates can be predicted with sufficient accuracy to allow prediction of non-volatile indoor PM2.5 ~ 10-20% precision. To produce a model for use in occupied buildings, further work capturing the effects of human occupancy on ventilation, penetration, deposition, and filtration may be required. Then, knowing regional characterizations of building leakage area, HVAC systems, and building operation, we expect that accurate estimates of the regional distribution of non-volatile indoor PM2.5 of outdoor origin will be possible. Inclusion of descriptions for these processes is underway.

Figure 3. Indoor/outdoor PM2.5 ratio calculated from size resolved data and predicted by



transient model. Upper panel compares measured ratio (dashed line, closed squares) against predicted ratio obtained with mid range model parameters and measured air exchange rate (solid line, closed diamonds) and predicted ratio obtained with deposition rate of 2.5 hr^{-1} (open diamonds). Note that a high deposition rate is necessary to approximately match the measured ratio. Lower panel (with expanded scale) shows predicted ratios obtained with mid range model parameters and measured infiltration rate (solid line, closed diamonds), mid-range model parameters and predicted infiltration rate (open diamonds), and upper estimate of deposition rate and measured infiltration rate (inverted triangles). Note that neither the over estimate of predicted infiltration rate nor the upper estimate of deposition rate produce more that 10-30% differences in predicted ratio.

CONCLUSION AND IMPLICATIONS

Measurements of air infiltration rate and size resolved fine particulate matter collected at a residence in the San Joaquin Valley, CA are compared with predictions from a building infiltration model and physically based transport model for non-volatile aerosol species. The results are considered in the context of producing a predictive model of indoor PM2.5 of outdoor origin that can be used at a regional level. The measured and predicted infiltration rates compare sufficiently well that the infiltration model can likely be used as part of an aerosol model. The aerosol transport model, however, does not capture the small indoor/outdoor PM2.5 ratios measured in the field. The reason for the discrepancy appears to be due to loss of volatile ammonium nitrate aerosol. Successful prediction of indoor PM2.5 in regions with significant concentrations of volatile aerosol, hence appears to require inclusion of physiochemical processes governing gas-particle phase changes.

Analysis and inclusion of the appropriate descriptions for these processes is currently being conducted.

ACKNOWLEDGEMENTS

The authors gratefully acknowledge the advice and assistance of Dr. William Riley, Dr. Iain Walker, and Darryl Dickerhoff. This research was supported by the Assistant Secretary for Fossil Energy, Office of Natural Gas and Petroleum Technology, through the National Petroleum Technology Office under U.S. Department of Energy Contract No. DE-AC03-76SF00098.

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INDOOR, OUTDOOR AND REGIONAL PROFILES OF PM2.5 SULFATE, NITRATE AND CARBON

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ABSTRACT
Fine particle concentrations were measured simultaneously at three locations: a regional monitoring site in Fresno, California, a backyard of an unoccupied residence in Clovis, California located 6 km northeast of the regional site; and indoors at the same residence. Measurements included 10-min determination of $PM_{2.5}$ nitrate, sulfate and carbon using an automated collection and vaporization system, and black carbon measured by light attenuation through a filter deposit. Specific outdoor $PM_{2.5}$ constituents were compared to assess the appropriateness of using regional data to model indoor concentrations from outdoor sources. The outdoor data show that, in general, the regional results provide a good representation of the concentrations seen at the building exterior. The indoor concentrations showed considerable attenuation as well as a broadening and time-lag for the concentration peaks. The concentration reduction was the largest for $PM_{2.5}$ nitrate, which appears to undergo phase changes in addition to indoor deposition and penetration losses.

INDEX TERMS
$PM_{2.5}$, nitrate, sulfate, black carbon

INTRODUCTION
Several epidemiologic studies have found a positive association between airborne particles and increased morbidity and mortality, even at the generally low levels of pollution (Dockery, et al., 1993, Pope, et al. 1995). Important to understanding these associations is assessment of human exposure to these particles. Since people spend most of their time indoors (Jenkins, et al., 1992, Robinson and Nelson, 1995), it is necessary to evaluate indoor exposures to outdoor particles with diameters below 2.5 µm ($PM_{2.5}$).

To better quantify indoor concentrations of outdoor $PM_{2.5}$, a detailed study is being conducted in an unoccupied home in the city of Clovis, California. Simultaneous, time-resolved, indoor and outdoor measurements of particle size distributions and chemical composition have been made to elucidate mechanisms of particle penetration, deposition (Thatcher et al., 2002) and phase transformations (Lunden et al., 2002). These analyses specifically address the dynamic nature of indoor/outdoor concentration relationships, and how the time-dependence of outdoor concentrations influence indoor concentrations. They explicitly examine the dependence on chemical composition, indoor – outdoor temperature relationships, and air exchange rates.

A related question is how well the concentrations measured inside the Clovis study home could be predicted on the basis of measurements at the central monitoring site, located 6 km distant. Important to this question is how well particle concentration data from the central monitoring site represent concentrations immediately outside the residence. This paper

examines the variability between the house and monitoring station, and its importance to predicting indoor concentrations.

METHODS

Measurements were made at a home (134 m^2) in Clovis, CA, as described by Thatcher et al (2002), and at the California Air Resources Board monitoring station in Fresno, as described by Watson et al (2001). The air monitoring station is located 6 km to the southwest of the study house.

At the study house, indoor and outdoor black carbon were measured with 20-min time resolution by attenuation of light through a particle deposit on a quartz fiber filter. Indoor and outdoor nitrate, sulfate, and carbon were measured with 10-minute time resolution using the integrated collection and vaporization method of Stolzenburg and Hering (2001) whereby particles are collected by humidification and impaction, and analyzed by flash-vaporization and quantification of the evolved vapor compounds. Nitrate concentrations are measured using low-temperature vaporization in a nitrogen carrier gas with assay of the evolved vapors using a chemiluminescent monitor equipped with a molybdenum converter. Sulfate and carbon analyses are performed using high-temperature heating, with analysis of the evolved sulfur dioxide by uv-fluorescence and carbon dioxide by nondispersive infrared absorption. Simultaneous indoor and outdoor measurements were obtained using a multicell system that shared a common set of gas analyzers. The sampling lines and cells for outdoor sampling were mounted within an enclosure through which outdoor air was drawn to maintain near-outdoor temperature at the point of sampling.

At the central monitoring site, replicate nitrate measurements were provided by two RP8400N monitors and sulfate measurements were obtained with a pre-release RP prototype (Rupprecht and Patashnick, Albany, NY). These are commercial instruments employing the same flash vaporization method used at the Clovis study house. Black carbon was measured with an Aethalometer (Andersen Instruments), which uses a similar measurement principle as the LBL black carbon instrument used at the study house. Integrated filter samples for chemical analyses were collected on selected days. Nitrates were measured on denuded, impregnated filters, sulfates on Teflon filters, and carbon on quartz filters.

The sample inlets at the residential site were located at a height midway between the house eves and the roof peak. This location was chosen to obtain samples which best represent the particle concentration at the building exterior. However, the chosen location is substantial lower and more sheltered than typical regional monitoring locations. At the central monitoring site, the sample inlets were mounted on the flat roof of a 2 story building and extended 2 m above the rooftop.

RESULTS

Quality Assurance: Time-integrated concentrations from the continuous measurements were compared to those obtained from the 12- and 24-hour filter samples at the Clovis study house and the Fresno central monitoring site. For nitrate, these comparisons yield $R^2>0.96$, with regression slopes of 0.88 and 1.14, respectively (Figure 1). The comparability of the indoor and outdoor cells was evaluated by sampling indoor air with both cells under high ventilation conditions. This was done over a 24-hr period on August 23rd, as shown in the first day of the time series plot of Figure 2. The two nitrate cells are correlated with an $R^2=0.97$. The indoor cell reported 15% lower values, on average, during this period.

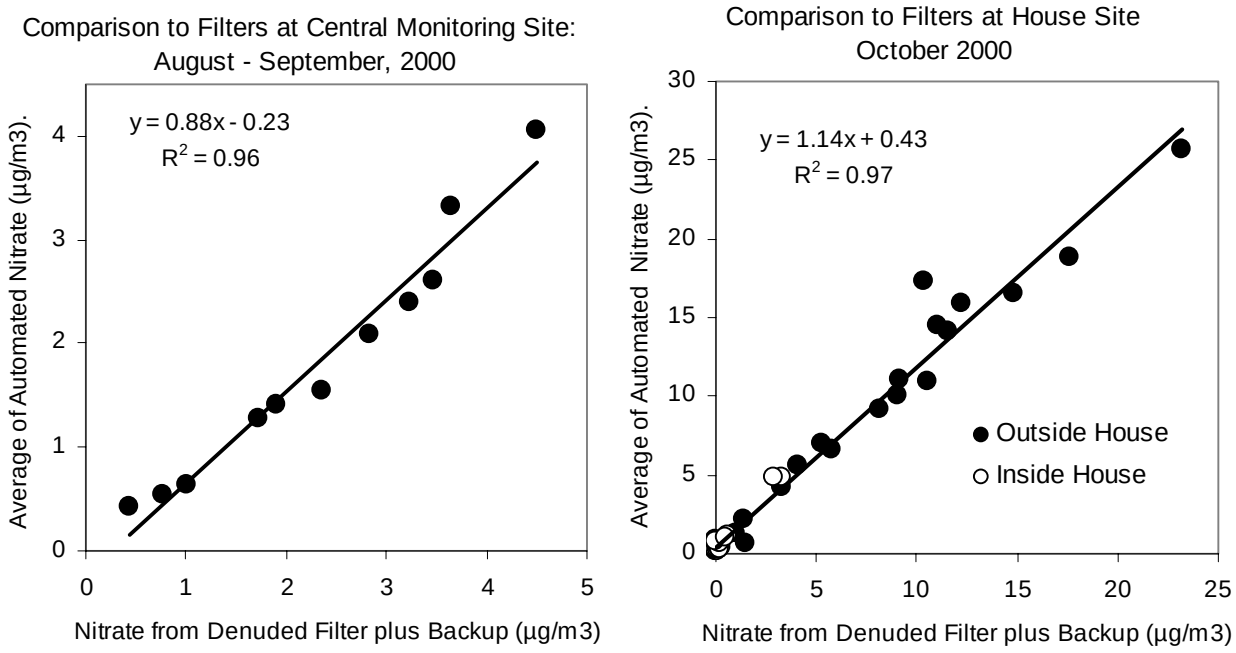

Figure 1. Comparison of automated nitrate measurements to parallel filter measurements (a) at the Central Monitoring Site in Fresno, and (b) at the study house in Clovis, 6 km to the northeast.

Daily Patterns in Nitrate Concentrations: An example of the daily variations in regional, outdoor and indoor nitrate concentrations is presented for the last four days of the time series in Figure 2 (August 24–27). Ambient concentrations exhibit a late morning to midday maximum, with 10-min peak concentrations reaching a factor of 3 or more higher than the daily average. On most days the concentrations at the study house were comparable to those at the central monitoring site, similar to that shown for August 24th. Occasionally the outdoor data showed excursions from the central site data, such as seen on August 26th. Notably, the nitrate concentrations inside the house are markedly lower than outside. These data were collected during our set-up period, when there was considerable activity in the house. Average temperature inside the house was warmer than outside by 7C.

Data collected during a two-week intensive study period in October 2000 are summarized in Table 1 below. Listed are the R^2 values (square of the correlation coefficients) and ratio of mean concentrations between measurements at the central monitoring site to those made immediately outside the study house, and between measurements inside the house and immediately outside. Correlations were computed for the 10-min data, and averages of these data to 6-hours. Indoor/outdoor correlations and ratios of mean values are also shown for the 12-hr filter based measurement. For the 10-min and 6-hr averaged nitrate data, the correlation between the central monitoring site and the outdoor measurement taken at the house exterior is much higher than for the comparison between inside the house and immediately outside. Both the automated nitrate system and the integrated filter data indicate mean indoor concentrations are less than 10% of the mean nitrate measured immediately outside during the same period.

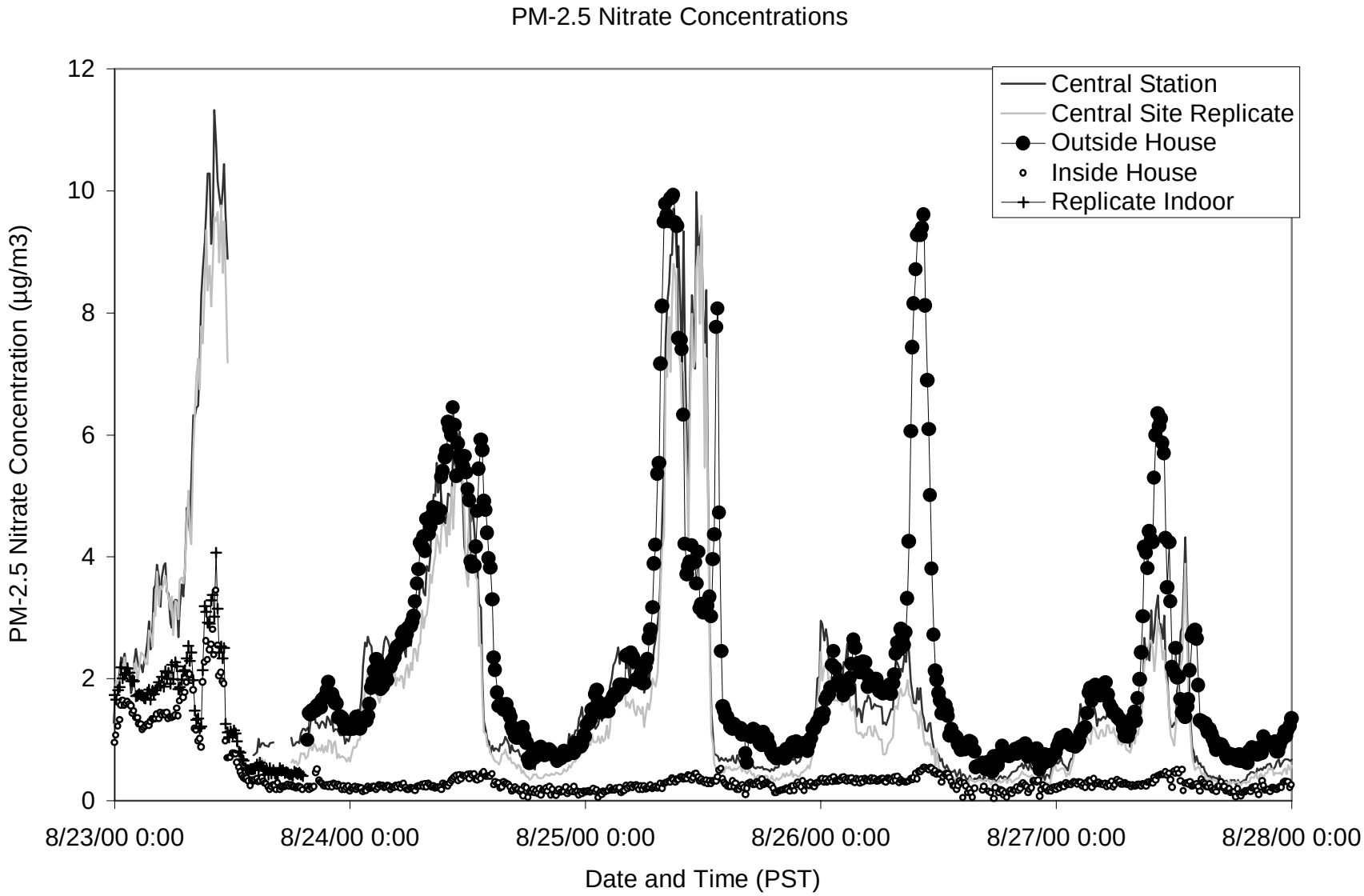

Figure 2. Example of $PM_{2.5}$ nitrate concentrations measured at central monitoring site and inside and outside of the Study House (see text).

Sulfate and Black Carbon Profiles: As with nitrate, the time variation and the concentration in $PM_{2.5}$ sulfate immediately outside the residence was comparable to that observed at the regional site. This is consistent with the regional nature expected for secondary constituents that are formed as a result of atmospheric oxidation processes. During the two-week intensive when the house was unoccupied, the mean of the indoor sulfate concentrations was approximately one-half of that outside. The correlation between indoor and outdoor measurements is higher than for nitrate, but lower than between outdoors and the central monitoring site. This is attributed to the time lag for infiltration into the building, whose the time constant varies with infiltration rate, and to the variability in depositional losses.

For black carbon, which is a directly emitted pollutant, we find similar patterns in ambient concentrations measured outside the house and at the central monitoring site. The black carbon concentrations were systematically higher at the central monitoring site (ratio of means = 1.7). Elevated black carbon at the central monitoring station is likely attributable to the closer proximity of traffic sources. Outside the house, local concentration peaks of black carbon were sometimes observed, likely originating from highly localized sources observed in the neighboring homes, such as wood burning and lawn mowing. These sharp black carbon peaks did not translate into indoor concentration peaks, presumably due to the short duration and small spatial extent of the carbon plumes, which may not have encompassed the entire building shell. As with the sulfate, correlation in concentrations between the central monitoring site and outside is greater than between indoors and immediately outside.

Table 1. Correlation and Ratio of Mean Values between Regional, Outdoor, and Indoor Concentrations for different averaging times and chemical constituents.

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Nitrate:	10-min Data	6-hr Average	12-hr Filter Data
R^2 for Replicates at Central Site	0.92	0.97	
R^2 for Outdoor vs Central Site	0.73	0.85	
R^2 for Indoor vs Outdoors	0.28	0.28	0.33
Regional Mean / Outdoor Mean	0.8		
Indoor Mean / Outdoor Mean	0.08		0.06

Sulfate:	10-min Data	6-hr Average	12-hr Filter Data
R^2 for Outdoor vs Central Site	0.81	0.93	
R^2 for Indoor vs Outdoors	0.61	0.44	0.33
Regional Mean / Outdoor Mean	1.2		
Indoor Mean / Outdoor Mean	0.53		0.51

Black Carbon:	20-min Data	6-hr Average	12-hr Filter Data
R^2 for Outdoor vs Central Site	0.53	0.73	
R^2 for Indoor vs Outdoors	0.26	0.55	0.72
Regional Mean / Outdoor Mean	1.7		
Indoor Mean / Outdoor Mean	0.51		0.62

CONCLUSIONS

By comparison to differences between indoor and outdoor concentrations, the concentrations of sulfate and nitrate immediately outside the house are well-represented by those measured at the central monitoring site. This is true even though the residential samples are taken in a , reduced height, sheltered location near the building in order to approximate concentrations at the building exterior. The similarities between the two sampling locations hold for both the average concentrations and the daily profile of concentration maxima. For black carbon, which is a primary pollutant with a large area source, the daily patterns are similar to the central site, but the absolute concentrations are lower. This indicates that a modeling effort that uses central monitoring data to represent concentrations immediately outside of homes should take into account the density and proximity of local area sources, such as traffic, in estimating these concentrations.

ACKNOWLEDGEMENTS

The authors dedicate this paper in memory of Dr. Joan Daisey, who played a key role in the initial phases of this project, and whose vision has guided our work. This research was supported by the Assistant Secretary for Fossil Energy, Office of Natural Gas and Petroleum Technology, through the National Petroleum Technology Office under U.S. Department of Energy Contract No. DE-AC03-76SF00098.

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PARTICLE PENETRATION THROUGH WINDOWS

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ABSTRACT
This study aims to characterize the fractional penetration of airborne particles through windows, one of the important sites of air leakage through building envelopes. Two aluminum windows were evaluated, one with weatherstripping and one without. For each experiment, a finished window was mounted and sealed in a plywood panel that separated two well-mixed compartments. A small pressure difference was established between the compartments to induce a constant rate of airflow through leakage paths in the window. Particles were injected into one chamber and their concentrations were measured in both chambers. Two methods were employed to evaluate the size-resolved particle penetration: a steady-state method and a dynamic, concentration growth method. The results indicate that airborne particles of 0.2 to 3 μm penetrate through both test windows fairly effectively, while significant particle losses are observed for particles smaller and larger than this range.

INDEX TERMS
Aerosols, Air pollutant transport, Particles, Pollutant penetration, Windows

INTRODUCTION
Windows are important contributors to air leakage in building envelopes. The investigation of air leakage through windows has been of interest owing to concerns such as reduced thermal comfort from cold drafts, increased energy consumption, and condensation problems. Less studied is the concern that air leakage through windows can also permit the penetration of ambient airborne particles into indoor environments, causing exposures that may have adverse health effects or contribute to material damage. For low-rise buildings, studies have indicated that most air leakage arises from openings in ceilings and walls; window and door components contribute about twenty percent to total air infiltration (Tamura, 1975; ASHRAE, 1993; Reinhold and Sonderegger, 1983). The extent of particle penetration through building cracks of well-defined geometry and through wall cavities has been modeled (Liu and Nazaroff, 2001). Experimental work using building-material cracks of idealized geometry has shown generally good agreement with model predictions (Liu and Nazaroff, 2002). However, for building components possessing complicated leakage paths, such as windows and other fenestration products, it seems more practical to develop an understanding of particle penetration by conducting experiments in the laboratory or in the field.

We know of no published study that evaluates the performance of windows with respect to the infiltration of ambient particles. In this study, particle penetration was measured in laboratory experiments for two windows. We believe that the methods used here can be applied to study particle penetration through other fenestration products (doors, curtain walls, etc).

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METHODS

Two operable, used aluminum sliding windows were obtained for the experiments; one is equipped with tubular gasket weatherstripping between moving sash and bottom frame (commercial class; designated as Wc), and the other is not weatherstripped (residential; Wr). Both windows have bristles between the sash and the frame to reduce air leakage. In addition, Wc has a wooden case that surrounds the aluminum perimeter frame closely. The commercial window was also tested with the joints between the wooden case and aluminum frame sealed by tape (test designated as Wc'). The frame sizes of Wr and Wc are 48.7 × 63.8 cm and 58.9 × 58.6 cm, respectively.

The finished window was mounted in a plywood panel so that all gaps between the window perimeter and the plywood were well sealed. Thus, the leakage paths within the window unit were the only air leakage pathways in these experiments. The window panel was inserted so that it separated the volumes of two identical plywood chambers (101.6 × 101.6 × 76.2 cm). A pressure difference (ΔP) of 1 Pa was created across the window by supplying air to chamber 1, some of which leaked into chamber 2. Both chambers were maintained at a net positive pressure to prevent uncontrolled particle infiltration from the laboratory. During the experiments, ΔP was monitored with a digital micromanometer (The Energy Conservatory, Minneapolis, MN, USA), which had been calibrated with a manometer (Microtector®, Model 1430, Dwyer Instruments Inc., IN, USA). A small fan was used to mix the air in each chamber.

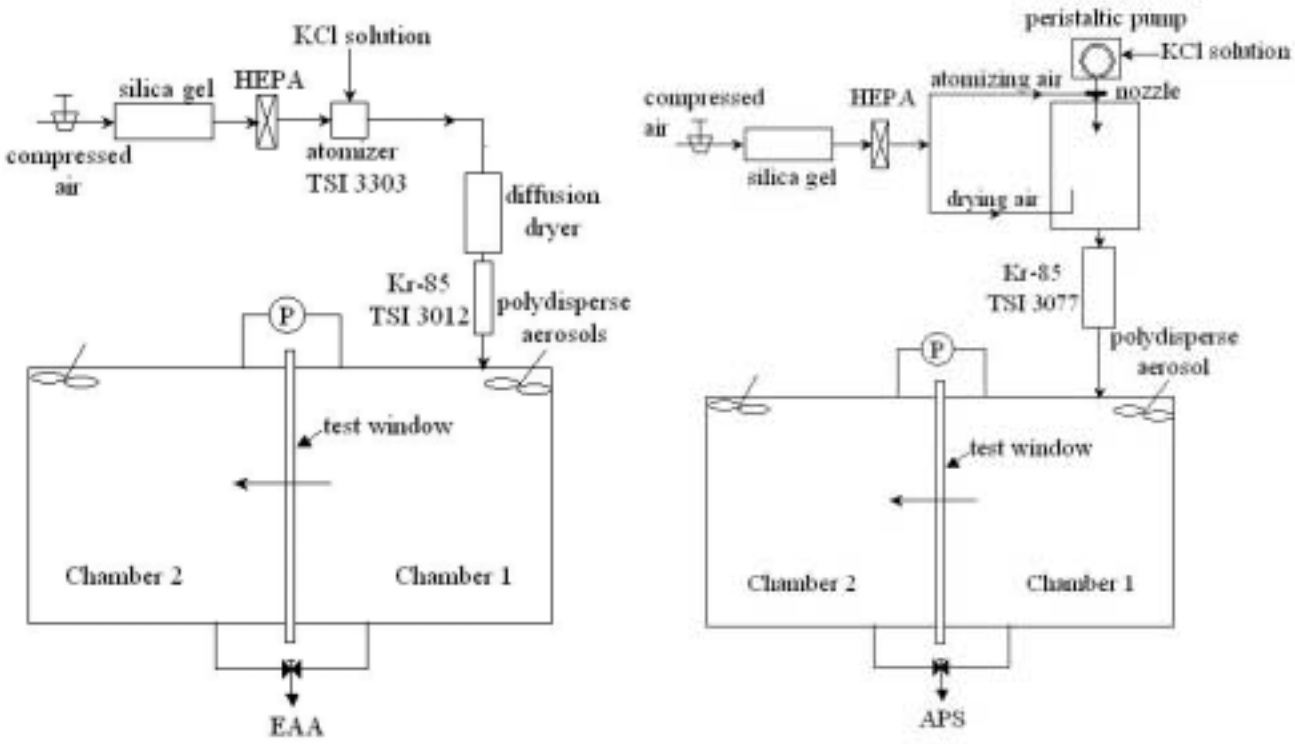
The experimental scheme involves continuously introducing polydisperse particles into chamber 1, and monitoring the concentration versus time in both chambers. The change of particle concentration in chamber 2 with time (dC_2/dt) can be represented by the following equation:

$$\frac{dC_2}{dt} = p\lambda_v C_1 - (\lambda_v + k_d)C_2 \qquad (1)$$

where C_1 and C_2 are the particle concentrations in chambers 1 and 2 (number cm^{-3}), p is particle penetration factor (dimensionless) through the window, and λ_v and k_d are air exchange rate (h^{-1}) and particle deposition coefficient (h^{-1}) in chamber 2, respectively. Note that C_1 and C_2 are measured as functions of particle diameter (d_p). It is evident from equation (1) that the particle penetration factor can be inferred from $C_1(t)$, $C_2(t)$, λ_v, and k_d once these parameters have been determined. Particle penetration is determined as a function of particle diameter through the appropriate application of equation (1) to experimental data.

Figure 1 shows the experimental schematics. After airborne particles were generated from atomizers (TSI 3075, St. Paul, MN; and a custom-built unit), they were dried and electrically neutralized prior to entering the chamber. Aerosol concentrations in both well-mixed chambers were continuously measured using an Aerodynamic Particle Sizer (APS, TSI 3320, St. Paul, MN) and an Electrical Aerosol Analyzer (EAA, TSI 3030, St. Paul, MN). Two sampling lines of identical length from chambers 1 and 2 met at a three-way valve, which was used to alternate the aerosol flow to the EAA or the APS. The air-exchange rate in chamber 2 was evaluated for each experiment by monitoring tracer gas (SF_6 or CO_2) concentration decay

with time, using a multigas monitor (Type 1302, Brüel&Kjær, Denmark) or a CO_2 monitor (Telaire 7001, Engelhard, USA).



(a) (b)

Figure 1. Experimental schematics for the generation and measurement of (a) submicron, and (b) supermicron particles.

RESULTS AND DISCUSSION

Particle loss rates

As shown in equation (1), air exchange and deposition onto chamber surface are the two particle removal mechanisms in chamber 2. To determine size-resolved particle loss rates under consistent airflow conditions, we performed a separate experiment that relies on measurement of size-specific, time-dependent particle concentration decay after a deliberate concentration increase. After particle concentration in chamber 2 was raised to a sufficient level, we stopped particle generation and monitored the concentration decay as particles were flushed out by particle-free air from chamber 1. Thus the overall particle loss rate ($\lambda_v + k_d$) can be determined by application of this equation:

$$\lambda_v + k_d = \frac{\ln(C_{2i}/C_{2t})}{t} \qquad (2)$$

where C_{2i} and C_{2t} are the initial and final particle concentrations measured in chamber 2, and t is the measurement time interval (h). The air-exchange rate was obtained by a similar method, except that the particle concentrations in equation (2) were replaced with tracer-gas concentrations.

Penetration factor

The particle penetration factor is defined here to be the fraction of particles that remain airborne as air enters chamber 2 from chamber 1 through the leaks of the test window. Two methods are employed to evaluate particle penetration as a function of particle size. The first method assumes that a steady-state condition prevails. The penetration factor is inferred by measuring the size-resolved particle concentrations in both chambers in response to a constant supply of polydisperse particles to chamber 1. Solving equation (1) for steady-state conditions, we have

$$p = \frac{\lambda_v + k_d}{\lambda_v}\left(\frac{C_2}{C_1}\right) \tag{3}$$

In the second method, the particle level in chamber 2 is first reduced to a negligible value by supplying particle-free air. Then, the increase of particle concentration is measured as polydisperse particles, supplied to chamber 1, penetrate through the window. We expect to see the growth of particle concentration in chamber 2 until it reaches the steady state, so we call the second approach the dynamic, concentration-growth method.

It is important to characterize the uncertainty associated with the experimentally determined penetration factors. A Monte-Carlo approach was applied to perform simulations, with the input parameters randomly sampled from normal distributions. The distribution means were designated as the experimentally determined values for air-exchange rate, particle deposition rate, and particle concentrations in both chambers, and the standard deviations were assigned so that the errors associated with the measurements were well described. To best evaluate the penetration factor from the concentration growth experiment, a least-square approximation was employed to fit the measured particle concentrations in chamber 2. In the study, thirty-two simulations were conducted for the uncertainty analysis in each experiment.

Figure 2 presents the calculated penetration factors from the simulations for Wr and Wc'. (The results for Wc agree closely with those for Wc'.) The solid symbols and the error bars indicate the average values of penetration factors and the ninety-five percent confidence interval, as determined by means of the dynamic, concentration growth method. The steady-state penetration factors, as determined from equation (3), are designated by open circles.

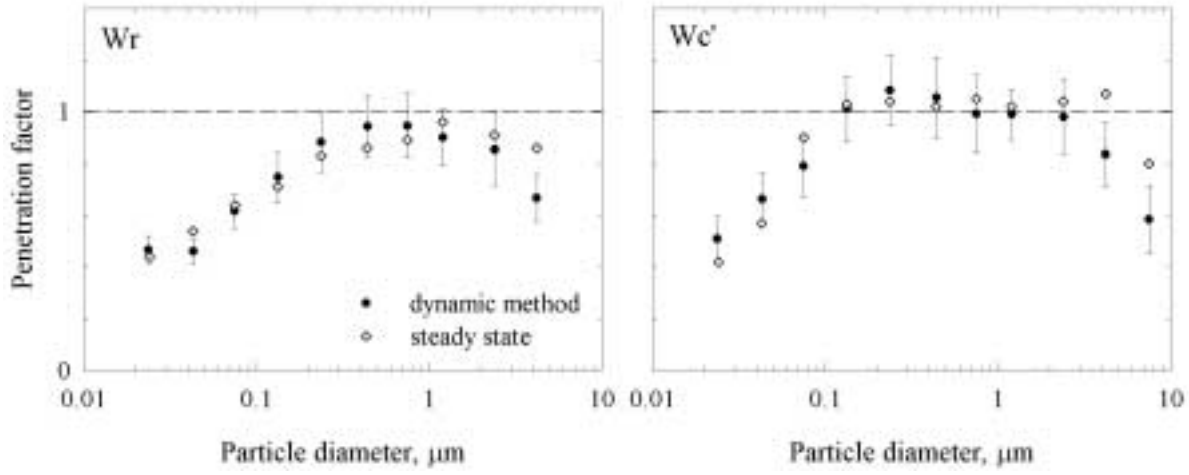


Figure 2. Particle penetration factors obtained for the two test windows from the steady state method, as well as the dynamic, concentration growth approach. Airflow through the window unit was induced by means of a steady pressure drop of 1 Pa.

As seen in Figure 2, particle penetration exceeds 80 % for 0.2-3 μm in Wr, while complete penetration is observed for 0.2-3 μm in Wc'. This indicates that the airborne particles in these size ranges penetrate through the windows fairly effectively. For particles larger or smaller than these sizes, significant particle losses arise, probably as a result of gravitational settling and Brownian diffusion, respectively (Liu and Nazaroff, 2001). For example, about 50% of 0.02 μm particles are lost from air leaking through these windows. In terms of experimental reliability, it is reassuring that the penetration factors estimated from the steady-state method agree moderately well with those that are determined from the dynamic, concentration-growth method for each test window.

For the smaller particles tested, the residential class window without weatherstipping appears to allow fewer particles to penetrate than the commercial class window with weatherstripping. This was confirmed by running a *t*-test, which revealed that the penetration factors for Wr were significantly lower than for Wc (at the 0.05 probability level) for particles smaller than 0.4 μm and larger than 2 μm. For particles between 0.4 and 2 μm, penetration through the two windows exhibits no significant difference statistically. These observations may result from different distributions of leakage-path sizes in the two windows.

The *t*-test was also used to compare the penetration factors of submicron particles for the commercial window with an unsealed frame (Wc) and a tape-sealed frame (Wc'); no significant difference was found. This indicates any additional air leakage between the aluminum perimeter and the wooden frame did not play a role in fractional particle penetration. (For experiments using supermicron particles, only Wr and Wc' were tested, since similar results were expected for Wc and Wc'.) Note that air flows through a variety of window leakage paths, which possess a distribution of geometrical dimensions. The overall particle penetration factor for a window unit is the flow-weighted-average penetration through each opening. Consequently, it is the distribution of window leakage dimensions that determines the overall performance of particle penetration, rather than the leakage area per se. Furthermore, since particle penetration also results from air infiltrating through leaks of window/wall joints and adjacent wall cavities, overall wall construction quality is expected to be a factor in particle penetration. Based on these insights, to minimize ambient particle penetration into buildings, improvements are needed in all elements: window design, manufacturing, installation, and maintenance. Reductions in particle penetration through building component systems, such as windows, can serve to reduce human exposure to ambient particles.

Window leakage

The notion of effective leakage area, used to evaluate the air tightness of building components, can be applied to characterize the windows tested in these experiments. The effective leakage area can be calculated from the following expression (ASHRAE, 1993):

$$A = \frac{Q}{C_d}\left[\frac{\rho}{2\Delta P}\right]^{\frac{1}{2}} = \frac{\lambda_v V}{C_d}\left[\frac{\rho}{2\Delta P}\right]^{\frac{1}{2}} \qquad (4)$$

where A is the effective (or equivalent) leakage area (m^2), ρ is air density ($kg\ m^{-3}$), Q is the air flow rate through the unit ($m^3\ h^{-1}$), C_d is the discharge coefficient for the leakage openings (dimensionless), and V is the chamber volume (m^3). The value of C_d is usually taken as 0.6 (as for a sharp-edged rectangular opening), though it might vary in the range of 0.6-1, depending on leakage characteristics (Heiselberg et al., 2000). Since the window perimeter is well sealed to the surrounding panel in the experiments, air leakage is expected to occur only through the sash/frame joints of the window assembly. The approximate effective leakage areas for Wr and Wc' at $\Delta P = 1$ Pa are 0.034 and 0.069 cm^2/lms (leakage area per linear meter of sash), respectively. These values are far less than the estimated effective leakage area (0.2 to 2.06 cm^2/lms) reported for single horizontal slider windows with weatherstripping (ASHRAE, 1993), even allowing for uncertainty in C_d and adjusting the reference pressure to 4 Pa. Such a discrepancy may be attributable to the fact that our experiments excluded air infiltration from wall/frame leaks. Extraneous leakage paths have been reported to contribute greatly to component air leakage (Carpenter, 1991; Louis and Nelson, 1995).

CONCLUSION

Laboratory experiments have been performed to investigate particle penetration through two windows. The penetration factors estimated from the steady-state method agree well with those determined from the dynamic, concentration-growth method. We have shown that more than 80% of particles in the diameter range 0.2-3 µm penetrate through either window. Penetration is lower for particles smaller or larger than this range. We point out that the overall particle penetration factor of a window assembly is determined by the distribution of leakage dimensions. Neither air-leakage area nor air-leakage rate, as aggregate terms that are commonly reported for assessing window air-tightness, are directly helpful in predicting fractional particle penetration. Although the small number of samples prevents us from drawing broad conclusions to apply to other window types, the results do provide some insight into expected values of particle penetration, especially when combined with our earlier modeling work (Liu and Nazaroff, 2001). Additional investigations along these same lines can improve our understanding of the factors that affect human exposure to particles of ambient origin. It is also conceivable that improved window assemblies could be developed that offer protection against exposure to ambient particles.

ACKNOWLEDGEMENT

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THE TRANSFORMATION OF OUTDOOR AMMONIUM NITRATE AEROSOLS IN THE INDOOR ENVIRONMENT

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ABSTRACT
Recent studies associate particulate air pollution with adverse health effects; however, the exposure to indoor particles of outdoor origin is not well characterized, particularly for individual chemical species. In response to this, a field study in an unoccupied, single-story residence in Clovis, California has been conducted. Real-time particle monitors were used both outdoors and indoors to quantify PM2.5 nitrate, sulfate, and carbon. The results show that reduced indoor sulfate and carbon levels are primarily due to deposition and penetration losses. However, measured indoor ammonium nitrate levels were often observed to be at significantly lower levels than expected based solely on penetration and deposition losses. The additional reduction appears to be due to the transformation of ammonium nitrate into ammonia and nitric acid indoors, which are subsequently lost by deposition and sorption to indoor surfaces. The size of the effect is dependent upon factors such as temperature, relative humidity, and ventilation rate.

INDEX TERMS
Aerosols, penetration, chemical transformation, ammonium nitrate, nitric acid, ammonia

INTRODUCTION
An understanding of the underlying reasons for the causes of the adverse health effects resulting from ambient particulate matter (PM) is of major scientific importance. These adverse health correlations are based upon data from outdoor regional monitoring sites (Dockery, et al., 1993, Pope, et al., 1995). However, individuals spend, on average, about 90% of their time indoors - 70% of that in homes (Jenkins, et al., 1992). This implies that indoor exposure to PM2.5 of outdoor origin may be an important element in determining the causes of these health effects.

To investigate the dynamics of indoor particles of outdoor origin, we performed a controlled series of intensive field experiments on an unoccupied, single-story residence in Clovis, California, a suburb of Fresno, in California's San Joaquin Valley. The conditions in the residence were manipulated to cover a wide range of conditions during several weeks of intensive measurements periods. The measurements focused on providing data on indoor and outdoor concentrations of PM2.5 particles as a function of chemical concentration under a variety of house ventilation, heating and cooling conditions. Indoor sources were minimized to enhance understanding the mechanism of the transport of outdoor particles indoors.

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METHODS

The experimental facility is a three-bedroom, single-story home (134 m^2) constructed in 1972. The house has standard height ceilings (2.4 m), a forced air heating and cooling system, and ceiling fans. The house is located in a residential suburb, surrounded by homes of similar size and mature trees. The flat topography of the area combined with the mature vegetative growth near the home resulted in low levels of wind loading around the building.

The chemical concentrations of indoor and outdoor aerosols were measured simultaneously with 10-minute time resolution using the integrated collection and vaporization cell (ICVC) system of Stolzenburg and Hering (2000). Particles were collected by humidification and impaction and analyzed by in-situ flash vaporization of the evolved vapor compounds. Nitrate concentrations were measured from the evolved vapors using a chemiluminescent monitor equipped with a molybdenum catalyst to convert higher oxides of nitrogen to nitric oxide. The sulfate quantitation was performed by analysis of the evolved sulfur dioxide by UV-fluorescence and total carbon was converted to carbon dioxide and analyzed by nondispersive infrared absorption. A four-cell ICVC system was used to perform simultaneous indoor and outdoor measurements; one pair for nitrate and one pair for a combined carbon and sulfate measurement. The system was located in the living room of the house, however the outdoor sampling cells were housed in an enclosure ventilated with outdoor air to maintain the system at outdoor temperature.

The important atmospheric gas phase species of ammonia, nitrous acid, nitric acid and sulfur dioxide were measured using an automated on-line ion chromatograph (IC) system. The IC system collects water-soluble gases using wet denuders, followed by concentration and ion chromatographic analysis based upon the technique of Buhr *et al.* (1995). The system utilized a dual-channel IC to enable simultaneous automated measurement of both anion and cation species. Denuders were located both inside and outside the house, and provided data with 30-minute time resolution.

Ventilation rates were measured using a continuous release SF6 tracer gas measurement system using a photoacousitic analyzer (Breul & Kajer, Model 1302). Temperature and relative humidity were measured both inside and outside the house using dual temperature and relative humidity probes (Vaisala HMD70Y) mounted in aspirated shielded enclosures.

Experiments were conducted during three intensive measurement periods; October 9-23, 2000, December 11-19, 2000 and January 16-23, 2001. During these intensives, entry into the house was limited to a one-hour period at midday to perform instrument checks and flow calibrations. Instrument control and data acquisition for all instruments were performed from outside the house, allowing for minimal disturbance of the indoor environment. Using the house as a laboratory, a range of conditions was explored during the intensives by manipulating the ventilation rate and indoor-outdoor temperature gradient through natural or mechanical means. The house experienced infiltration rates in the range of 0.2 to 0.5 air changes per hour (ACH) when allowed to operate under naturally occurring conditions. When the doors and windows were opened, the ventilation rate could increase up to 1 ACH. In order to further expand the experimental parameter space to a regime where the residence time for air in the house approached the time required for other processes, such as deposition, to occur, we utilized mechanical

ventilation techniques to achieverates in the range of 2 to 6 ACH. The forced-air heating/cooling system was used to provide changes in the indoor/outdoor temperature differential in addition to conditions of either no, continual, or intermittent fan operation.

RESULTS AND DISCUSSION

Figure 1 shows time series of concentrations for measured total carbon, sulfate and nitrate from the ICVC system during the December intensive measurement period. Figure 1 also shows the ventilation rate measured during the same period. Note that the rate measured during 12/16 PM, 12/17 AM, 12/28 PM and 12/19 AM reflect the high values achieved using mechanically driven ventilation. What is immediately apparent is that there is a great deal of variability in both the indoor and outdoor concentration measurements for these three species. The difference between the indoor and outdoor concentrations is also highly variable. In general, however, the difference between the indoor and outdoor concentration decreases appreciably during periods of high air change rates. This result is intuitive as the characteristic time for particle gain by penetration is significantly greater than that for loss by deposition at these high ventilation rates. Of the three particulate species shown in Fig. 1, sulfate is generally considered to be the most stable upon transport from outdoors to indoors. A forward marching fit of the sulfate data using the time-dependent physical mass balance model results in values of 0.95 for the penetration factor and 0.19 hr^{-1} for the deposition loss rate, both of which are reasonable (Thatcher, et al, 2002).

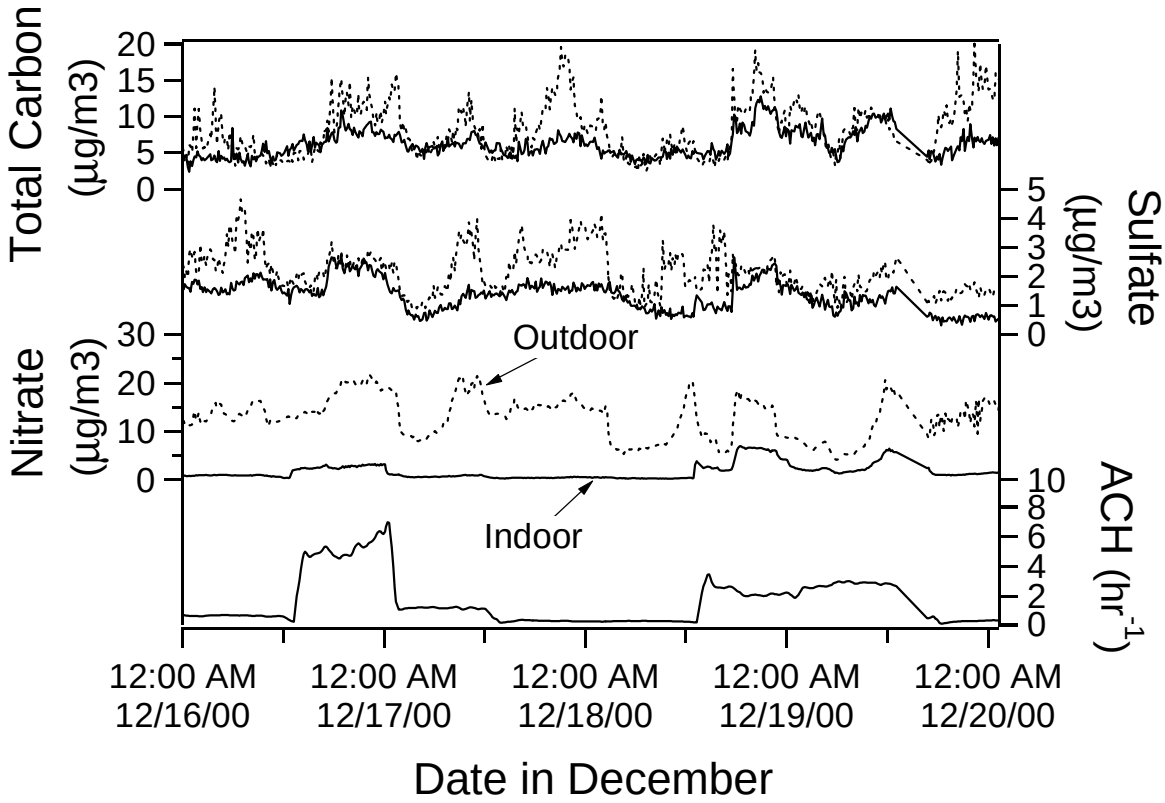

Figure 1. Data from the ICVC system showing the variation in indoor (solid line) and outdoor (dashed line) carbon, sulfate, and nitrate for a four day period during the December intensive. The lowest plot on the diagram shows the ventilation rate as a function of time.

The differences between outdoor and indoor nitrate particle concentrations are significantly larger than those observed for the other particle species. The indoor concentrations are most often quite low, rising to appreciable levels only during periods of high air change rates. Ammonium nitrate is volatile, and the gas-to-particle partitioning is highly dependent on factors such as temperature, relative humidity and the gas phase concentrations of ammonia and nitric acid as outdoor air is transported indoors. Previous experimental investigations have demonstrated that ammonium nitrate aerosol undergoes phase transitions during transport into houses (Hering and Avol, 1996). During the winter period, the nitrate aerosol moved from a cooler outdoor to a warmer indoor environment, shifting the equilibrium of the nitrate formation reaction,

$$HNO_3\ (g) + NH_3\ (g) \leftrightarrow NH_4NO_3\ (aerosol) \qquad (1)$$

to the left towards the gas phase. The gases diffuse toward and absorb to surfaces inside the house, further driving the equilibrium toward the gas phase. A final, and important factor affecting equilibrium is how quickly either gas is lost to surface uptake.

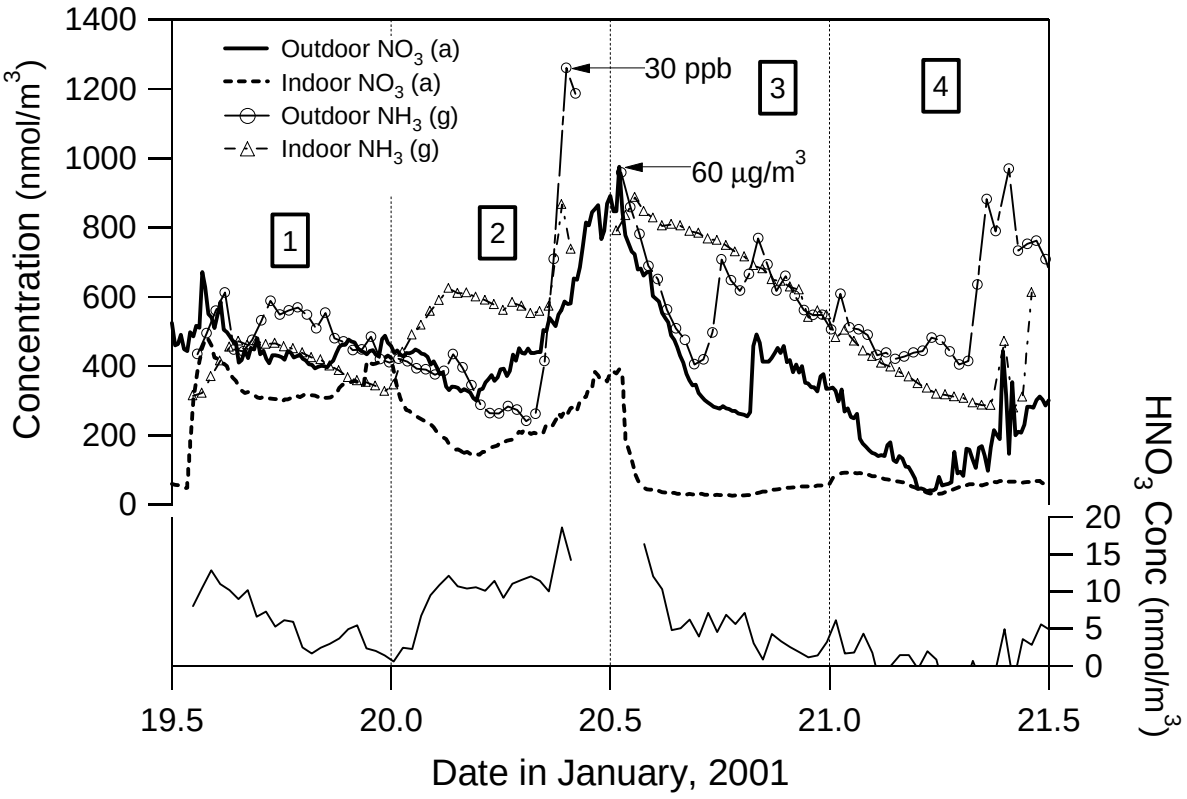

Figure 2. High time resolution measurements of the concentration of ammonium nitrate particulate (NH_4NO_3) and gaseous ammonia (NH_3) measured outside and inside the residence. The lower portion of the graph shows the concentration of the gaseous nitric acid (HNO_3) inside the residence. The numbered periods are described in the text.

The importance of the ammonium nitrate equilibrium can be seen in Fig. 2, which shows results of the ICVC system for nitrate and the automated IC system for ammonia and nitric acid for four different 12 hour periods during the January intensive. The indoor environment for each 12 hour period was manipulated as follows: (1) ventilation rate of approximately 4 h^{-1} with the heat off, (2) ACH ~5h^{-1} with the heat on at 20 C, (3) ACH

~0.3 h^{-1} with the heat off, and (4) ACH ~1hr^{-1} with the heat off. Note how well the outdoor ammonia (NH_3) and nitrate (NH_4NO_3) measurements are correlated, particularly on Jan 20th. During the period from noon on January 19th to noon on Jan 20th, the indoor ammonia and indoor nitrate are anticorrelated – as the nitrate concentration decreases, the ammonia increases. Indeed, throughout the entire measurement campaign, the indoor ammonia levels were often higher than those found outdoors due to this dissociation. During periods (1) and (2), the characteristic time for nitrate penetration from outdoors was large enough to balance the amount lost to dissociation, resulting in relatively large indoor nitrate levels. This period was one of the few times when significant levels of indoor nitrate were observed. The addition of heat to the house between conditions (1) and (2) is enough to cause a perceptible shift towards the gas phase. When the air change rate was drastically lowered between conditions (2) and (3), much of the nitrate dissociated, resulting in large indoor ammonia levels.

Throughout the entire experiment, the nitric acid (HNO_3) concentration indoors was low to undetectable. Based on Eq. (1), equal amounts of ammonia and nitric acid should be formed, however the data show a large difference between the concentrations of these two gases. After dissociation, the nitric acid gas is sorbing to the walls at faster rates than for the ammonia gas, which appears to accumulate. We attribute this to acid-base chemistry causing the nitric acid to be more tightly bound to the walls. The loss of nitric acid serves to continue to drive the equilibrium toward the gas phase.

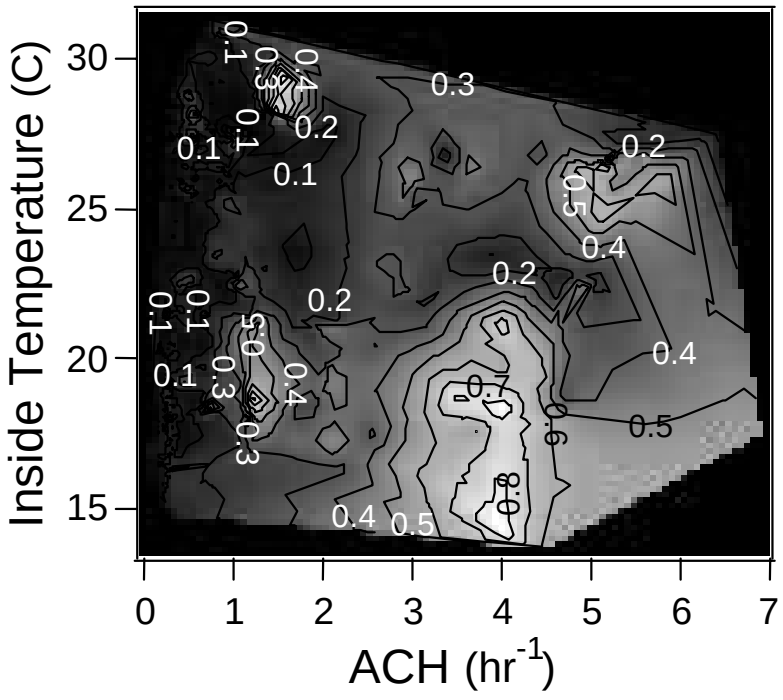

Figure 3. Contours of the ammonium nitrate indoor/outdoor ratio as a function of indoor temperature and ventilation rate.

The fate of ammonium nitrate aerosol once transported into the house is a complex function of the ventilation rate, the constituents in the gas phase, and the temperature. The conditions that result in high nitrate can be seen more clearly in Fig. 3, a contour plot of the indoor/outdoor ratio of ammonium nitrate as a function of both indoor temperature and ventilation rate. Two conditions result in increased indoor nitrate

concentrations. In the upper left of the figure, the house was open during the fall intensive. In this case, the indoor and outdoor environments were similar and the ventilation rate was relatively large. Thus, the rate of nitrate loss due to dissociation was smaller than the supply of nitrate due to penetration from outdoors resulting in increased indoor concentrations. The lower center of the graph shows the elevated winter conditions. Air change rates were very large and the temperature was low due to forced ventilation. The large air change rate serves to constantly replenish the ammonium nitrate lost to dissociation and deposition

CONCLUSIONS

A series of simultaneous indoor and outdoor measurements were performed at a residence, providing real-time, chemically speciated data for the significant aerosol species of nitrate, sulfate, and carbon as well as the gas species ammonia and nitric acid. The highly time resolved data show considerable variability in both the outdoor and indoor concentrations of PM2.5 constituents. This results in a wide range of ratios of indoor to outdoor concentrations that might not be evidenced in lower time resolved measurements. More importantly, the dissociation of the nitrate aerosol illustrates that an exposure assessment based on total particle mass measured outdoors may obscure the actual causal relationships, especially for indoor exposures to particles of outdoor origin. Nitrate is a significant outdoor pollutant in the Western United States. The extent to which it may or may not be a significant source of indoor exposure has important policy implications for control of sources that lead to nitrate formation. These results emphasize the need for chemical characterization of particulate, and further studies of the physical and chemical transformation processes that influence the indoor concentration of particles of outdoor origin.

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The authors dedicate this paper in memory of Dr. Joan Daisey, who played a key role in the initial phases of this project, and whose vision has guided our work. This research was supported by the Assistant Secretary for Fossil Energy, Office of Natural Gas and Petroleum Technology, through the National Petroleum Technology Office under U.S. Department of Energy Contract No. DE-AC03-76SF00098 and the Western States Petroleum Association.

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FACTORS AFFECTING THE CONCENTRATION OF OUTDOOR PARTICLES INDOORS: EXISTING DATA AND DATA NEEDS

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ABSTRACT
Accurate characterization of particle concentrations indoors is critical to exposure assessments. It is estimated that indoor particle concentrations depend strongly on outdoor concentrations. For health scientists, knowledge of the factors that control the relationship of indoor particle concentrations to outdoor levels is particularly important. In this paper, we identify and evaluate sources of data for those factors that affect the transport to and concentration of outdoor particles indoors. To achieve this goal, we (i) identify and assemble relevant information on how particle behavior during air leakage, HVAC operation, and particle filtration effects indoor particle concentration; (ii) review and evaluate the assembled information to distinguish data that are directly relevant to specific estimates of particle transport from those that are only indirectly useful; and (iii) provide a synthesis of the currently available information on building air-leakage parameters and their effect on indoor particle matter concentrations.

INDEX TERMS
Exposure assessment, indoor environment, mass-balance model, particles, air infiltration

INTRODUCTION
Because the US population is estimated to spend on average more than 80% of their time indoors, accurate characterization of particle concentrations indoors is critical to assessing total exposure to particles. In most cases, concentrations of ambient particles indoors depend primarily upon the quantity of particulate matter that penetrates through the building shell or is transported via the heating, ventilation, and air conditioning (HVAC) system. In addition, the concentration of particles indoors is affected by several other factors such as filtration, ventilation, deposition, re-emission, and indoor sources. To evaluate and model human exposures to particles requires information on these characteristics. We refer to such factors as Concentration-of-Outdoor-Particles-Indoors (COPI) factors. Most COPI factors vary from one type of building to another. Thus, COPI factors cannot be represented by a single value. Instead the variation in these factors needs to be explored and recorded. How this variation relates to building design and operation also needs to be evaluated. In this paper, we identify the COPI factors that are most needed for assessing human exposure to outdoor particles indoors and then compile and evaluate information sources for these COPI factors.

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We consider four categories of COPI-factor information in our analysis. First is the nature of building shell leaks and what influence they have on airflow and particle transport indoors. Second is the role of HVAC systems in supplying air to the building interior, typically through filter media of varying types and filtration efficiency. Third is information obtained from experiments in which indoor/outdoor particle concentrations are measured and particularly whether there is ancillary information about the building and/or its leakage characteristics. Fourth is information from papers and reports in which particle penetration is estimated and discussed. We address these issues through three tasks that are summarized in the section below on methods.

METHODS

Considerable previous work has been expended to understand and measure building leakage and other airflow characteristics as they affect air infiltration and ventilation. However the connection between these airflow pathways and particle entry into buildings remains poorly understood. To address this issue, we examined a number of experiments and models to better characterize the transport of ambient particles from outdoor air to the indoor environment. We address these issues through three tasks—information retrieval, information evaluation, and information synthesis.

Task 1, Information Retrieval

We first identified relevant information, databases, etc. needed to assess how transport of particles from outdoor to indoor air is impacted by leaks, HVAC systems, particle mass-balance data, and penetration data. In the US, key sources of information for COPI factors include the US Department of Energy (DOE) Residential Energy Consumption Survey (RECS) (US DOE, 2000), the DOE Commercial Building Energy Consumption Survey (CBECS) (US DOE, 1997, 1998), the US Department of Commerce (1999) surveys of housing characteristics, and the US EPA Building Assessment Survey and Evaluation (BASE) study (US EPA, 1994). In addition to these surveys we had access to databases at LBNL and made use of summary data available in the literature.

Task 2, Information Evaluation

Once the relevant information was assembled, our next task was to review and evaluate this information to determine which resources provide relevant COPI factors. In this task, we used three criteria by which we evaluated the available information. First was the quantity of information available. For example, is the information derived from a large survey or from a few test houses? Second was the quality of the information. For example, the reliability of the methods used—direct measurement, modeling, replication, quality control, etc. Third was the representativeness of the available information. For example how well are the population of residential and commercial buildings represented, and were the samples stratified so as to properly capture variability by geographic region, season of the year, building age, etc. We used a review and evaluation of currently available information to identify data needs, current resources, and data gaps for COPI factors.

Task 3, Information Synthesis

Our third task provides a synthesis of the currently available information on COPI factors and their effect on particle concentration indoors. We used this effort to identify areas for which there are important gaps or weaknesses in the available information, particularly as it relates to linking air leakage, particle transport, and particle fate.

RESULTS

Based on the methods described above we have identified and ranked those relevant factors that affect the concentration of outdoor particulate matter indoors. The relevance of different factors in controlling the concentration of outdoor particles indoors is established in the context of an indoor/outdoor particle mass balance model. The concentration of outdoor particles in the indoor environment is a balance between the rates at which outdoor particles enter and leave the air within the building and the rates at which they are removed, transformed, and re-emitted in the indoor environment. In general, a mass balance equation representing the change in indoor concentration, C_i, within the volume, V, over time, t, can be written as follows:

$$\frac{dC_i V}{dt} = [\text{Entry of Outdoor Particle Mass}] - [\text{Exit of Indoor Particle Mass}] - [\text{Mass Removal Indoors: deposition, filtration, transformation}] + [\text{Mass Re-emission Indoors: resuspension, transformation}] \quad (1)$$

In Figure 1, we illustrate the processes that affect the transport and concentration of indoor particles outdoors.

A Set of Relevant COPI Factors

Particle entry rates are influenced by the overall ventilation rate, the route of entry (e.g., through filters, cracks, windows), and particle penetration efficiencies. While considerable work has been expended on defining the factors affecting ventilation rates, there is still significant uncertainty concerning the route of entry in several areas, such as crack size distributions or infiltration rates in large commercial buildings. There is even more uncertainty in the area of particle penetration efficiency. Removal mechanisms

Figure 1. Transport and transformation mechanisms that define particulate matter concentrations in the indoor environment.

and rates (e.g., deposition, filtration, and exfiltration) are probably the best understood of the mass balance components, with experimental data and well formulated theoretical models for at least some of the components. However, variability among buildings is very high and the causes for this variability are poorly understood. Chemical transformation in the indoor environment is only just beginning to be investigated and could prove to be an important loss factor for some particle types. It is also a potentially large confounding factor in that some transformations produce more particles or change the size distribution of particles.

The construction of a building and the operation of building HVAC and other systems will affect the overall ventilation rate, as well as the routes through which air enters a building. The following important building characteristics were identified for their effect on particle entry rates into buildings:

measured or predicted ventilation rates
window opening,
prevalence of mechanical ventilation
air leakage in ducts,
pathways for air and particle leakage into buildings,
particle penetration factors, and
stairwells and buoyancy induced flow.

Particle removal in buildings may be due to intentional removal mechanisms (such as filtration with the HVAC system) or inherent removal mechanisms (such as particle deposition to surfaces). We have identified the following factors for their significance in impacting loss and persistence of particles transported from outdoor air indoors:

filter efficiencies,
rates and times of air flow through filters,
particle removal by auxiliary air cleaners,
particle deposition rates in buildings, and
resuspension factors.

Evaluation of Available Data on COPI Factors

The DOE RECS is an ongoing survey of energy use in residential houses and goes back to 1978 (US DOE, 2000). It is a national statistical survey, sampling approximately 5,900 housing units in the 1997 survey alone. Surveys are based on either 30-minute personal interviews, telephone interviews, or mailed questionnaires. The resulting data describes energy use and many characteristics of the United States housing stock including the size of housing units, types of heating and cooling systems installed, and numbers of household members present. The survey is also a detailed source of information for quantifying the status and types of residential buildings geographically. The DOE, Energy Information Administration's CBECS is a periodic survey of energy use in commercial buildings. From this survey, DOE has issued two major reports, the first on energy consumption (US DOE, 1998) and the second on commercial building characteristics (US DOE, 1997). CBECS is a national statistical survey comparable to the RECS in its completeness and characterization of energy consumption in commercial buildings. Relevant information in the CBECS survey includes data on types, numbers,

and sizes of buildings, HVAC system characteristics, and presence or absence of operable windows.

The US Department of Commerce (1999) also conducts surveys of housing characteristics using interviews. The 1997 survey of 56,000 housing units in 46 metropolitan areas collected data on single-family homes, apartments, and mobile homes. The most relevant data from this survey appear to be basic housing characteristics such as age, fuel use, heating and air conditioning equipment, and number of occupants. The EPA BASE study (US EPA, 1994) was carried out to obtain baseline information on indoor air quality, health symptom prevalence, and relevant building characteristics in US office buildings. One hundred office buildings were studied for the period 1996 to 1998. Relevant data from the BASE study include measured indoor and outdoor PM2.5 and PM10 concentrations, temperatures, and humidity levels; descriptions of the building envelope; descriptions of interior finish materials (which may affect particle deposition); a detailed description of the buildings' HVAC systems, including information of the types of particle filters (manufacturer, model, and usually an efficiency rating); and rated and measured air flow rates in HVAC systems. These data apply to a test space within each building, which is often smaller than the entire building. The BASE Study emphasizes large office buildings with mechanical ventilation and air conditioning.

Data Gaps and Data Needs for COPI Factors

Data quality, quantity, and representativeness were evaluated for key factors such as HVAC design and operation, infiltration and leakage area, deposition, penetration factors, and natural ventilation. We found that none of the critical factors affecting the indoor/outdoor particle relationship are yet well enough characterized to provide reliable inputs to exposure models. Particle deposition is an important factor affecting indoor particle concentrations in all types of buildings. However, deposition loss rates appear to be highly sensitive to indoor environmental conditions and particle characteristics, leading to significant variations in experimentally determined deposition rates. Another critical factor that is inadequately understood is the fraction of outdoor particles that remain in infiltrating air and enter the building interior, typically referred to as the penetration factor. Information of the efficiency of various types of filters is fairly high, both in quality and quantity. However, data on the types of filters used in residences, on time-average air flow rates in central forced-air residential heating and cooling systems, and on the use of auxiliary filtration systems are very sparse, leading to large uncertainties in particle filtration rates in residences. The extent to which windows are opened is another poorly understood factor that has a significant impact on indoor concentrations of outdoor particles. The available information on particle resuspension rates is extremely limited. There are still significant gaps in the assessment of factors such as infiltration rates into commercial buildings, the prevalence of local exhaust use in residential buildings, and the particle penetration data across a variety of buildings.

DISCUSSION

The concentration of outdoor particles in the indoor environment is a balance between several competing processes: particle entry, particle removal, and particle re-emission. Of these three processes, the least is known about particle re-emission. As a consequence re-emission rates introduce a high degree of uncertainty for predicted particle concentrations in occupied spaces. In contrast to COPI factors that are needed to

carry out mass balances on particles, the quantity and quality of data on basic building characteristics is quite high for both residential and commercial buildings. A more detailed presentation of the methods and results discussed in this paper is available in recent report by Thatcher et al. (2001) which is available from Lawrence Berkeley National Laboratory.

CONCLUSION AND IMPLICATIONS

Our review and evaluation affirms that understanding the processes affecting the concentration of outdoor particle indoors is essential for improving assessments of human exposure to particles of outdoor origin. This is particularly important for assessors who need to consider the spatial and temporal variation of indoor exposures to outdoor particles. Based on a detailed review and evaluation of data available and needed to link indoor concentration of particles to outdoor concentrations, we have determined that the most critical missing information includes the following:

measured particle penetration factors,
measured particle deposition rates in commercial and institutional buildings,
infiltration rates in commercial buildings,
the types of filters used in all buildings (except large offices),
window opening behaviors, and
rates of indoor resuspension of particles transferred indoors from outdoors.

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THE EFFECT OF PENETRATION FACTOR, DEPOSITION, AND ENVIRONMENTAL FACTORS ON THE INDOOR CONCENTRATION OF PM2.5 SULFATE, NITRATE, AND CARBON

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ABSTRACT
Indoor exposure to particles of outdoor origin constitutes an important exposure pathway. We conducted an intensive set of indoor particle measurements in an unoccupied house under differing operating conditions. Real-time measurements were conducted both indoors and outdoors, including PM2.5 nitrate, sulfate, and carbon. Because the time-scale of the fluctuations in outdoor particle concentrations and meteorological conditions are often similar to the time constant for building air exchange, a steady state concentration may never be reached. The time-series experimental data were used to determine the effect of changes in air exchange rate and indoor/outdoor temperature and relative humidity differences on indoor particle concentrations. A multivariate regression was performed to investigate the difference between measured indoor concentrations and results from a simple time-dependent physical model. Environmental conditions had a significant effect on indoor concentrations of all three PM2.5 species, but did not explain all of the model variation.

INDEX TERMS
Particle, deposition, penetration factor, ammonium nitrate, ammonium sulfate, carbon

INTRODUCTION
Particulate air pollution is associated with increased morbidity and mortality even at the generally low levels of pollution in United States cities (Samet, et al., 2000). The exact compounds and/or particle size ranges responsible for these health effects have not yet been determined. Exposures to particles of outdoor origin which occur while indoors may constitute a significant fraction of the overall exposure to hazardous particles since typically people spend up to 90% of their time indoors (Jenkins, et al., 1992, Robinson and Nelson, 1995). Indoor concentrations of particles of outdoor origin can be on the same order as outdoor concentrations (Ott et al., 2000; Riley et al., 2001).

Separating the effects of deposition and penetration in a full-scale house is difficult. In this study, we measured the indoor and outdoor concentrations of PM 2.5 nitrate, sulfate, and carbon over a period of several days. A simple time dependent physical model is then used to determine the penetration factor, defined as the fraction of particle in the infiltrating air which pass through the building shell, and deposition loss rate for each compound. The difference between the modeled and measured values is used in a multiple variable regression to examine the effect of changes in temperature, relative humidity, and air exchange rate on model fit.

METHODS

The experiments were performed in a 134 m^2 home in Clovis, California. The home was constructed in 1972, with a stucco exterior and sliding aluminum frame windows. The house is single story, with standard height ceilings (2.4 m), a forced air heating and cooling system, and ceiling fans (which were operated during the experiments to promote mixing). An additional oscillating fan was operated in the living room to disperse tracer gas and promote mixing near the particle measurement equipment. The building is located in a residential suburb, surrounded by mature trees and homes of a similar height and size. The flat terrain and high level of sheltering resulted in relatively low wind speeds near the building.

PM2.5 nitrate, carbon and sulfate were measured with 10-minute time resolution using the integrated collection and vaporization cell (ICVC) method of Stolzenburg and Hering (2001). This method collects PM2.5 particulate matter by humidification and impaction onto a 1 mm diameter spot on a metal substrate. The sample is then analyzed by flash-vaporization and quantitation of the evolved vapor compounds. Nitrate concentrations are measured using low-temperature vaporization in a nitrogen carrier gas with quantitation of the evolved vapors using a chemiluminescent monitor equipped with a molybdenum converter to reduce higher oxides of nitrogen to nitric oxide. Sulfate and carbon analyses are performed using high-temperature heating, with analysis of the evolved sulfur dioxide by UV-fluorescence and carbon dioxide by nondispersive infrared absorption. Indoor and outdoor measurements were performed simultaneously using a four-cell system. One pair of cells was used for nitrate measurements. A second pair was used for the combined measurement of carbon and sulfate. The nitrate and sulfate-carbon cells used to measure outdoor concentrations were housed indoors inside a box that was ventilated with outdoor air to maintain near-outdoor temperature at the point of sampling. Outside air was transported into the system through a large inlet tube, with the sample drawn through an isokinetic inlet in the center of the tube. Air infiltration rates for the house were continuously measured using a constant injection of sulfur hexafluoride tracer gas and measurement with an photo-acoustic detector system.

For our analysis, we used a simple physical mass balance equation, reduced to include only the effects of deposition and penetration losses:

$$\frac{\partial C_I}{\partial t} = C_O P \lambda_v - C_I(\lambda_v + \beta) \qquad (1)$$

Where: C_I = indoor particle concentration at time t (# cm^{-3}),
t = time (h^{-1}),
C_o = outdoor particle concentration at time t (# cm^{-3}),
P = penetration factor,
λ_v = air exchange rate (h^{-1}), and
β = deposition loss rate (h^{-1}).

This equation is solved fairly easily for a constant outdoor concentration and stable infiltration rate. Unfortunately, the system may not reach steady-state conditions, so we solve the equation using a basic 'forward-marching' scheme with time step, Δt, as shown below:

$$C_i(t_2) = C_i(t_1) + PC_o(t_1)\lambda_v(t_1)\Delta t - C_i(t_1)(\lambda_v(t_1) + \beta)\Delta t \qquad (2)$$

In these experiments, the time step used was 10 minutes, corresponding to the measurement interval. The deposition loss rate and penetration factor were adjusted to minimize the squared relative errors summed over time, where the squared relative error is defined as the square of the difference between the measured and modeled indoor concentration divided by the measured concentration. This weighting scheme gives equal weighting to relative errors in predictions at both high and low concentrations. The Excel spreadsheet function SOLVE was used to determine the deposition loss rate, β, and penetration factor, P, which minimized the sum of the squared relative errors for each chemical compound.

The outdoor concentrations and infiltration rates entered into the modeled are a time-series of measurements. Variations in these two parameters are expected to have the largest effect on indoor concentration. The model determines the constant deposition loss rate and penetration factor which best fit the data. However, the deposition loss rate and penetration factor may vary with time due to changes in the indoor environment, such as temperature or relative humidity. In addition, the particle size distribution for a PM 2.5 class may change over time, leading to further changes in the deposition and penetration rates. Further analysis is required to assess the importance of these effects on indoor concentrations. The model also assumes that there are no other significant loss or gain mechanisms affecting indoor particles. Volatilization, hygroscopic growth or loss, phase change, or other processes could affect the indoor concentration of PM 2.5 sulfate, nitrate, or total carbon.

To investigate the influence of some of these confounding effects, a multivariate linear regression was performed on the correlations between the model/measurement difference at each point in time and indoor/outdoor relative humidity and temperature differences, air exchange rate, and outdoor concentration.

RESULTS

Figure 1 compares time series of the modeled and measured indoor concentration for each compound. During four 12 hour periods (1/19 PM, 1/20 AM, 1/21 PM, and 1/23 AM), the house was pressurized with outdoor air via a large fan mounted in a windows. During these periods, the penetration factor was set to 1, indicating an assumption of no significant particle losses within the fan. During six 12 hour periods (1/17 AM, 1/18AM, 1/18PM, 1/21 AM, 1/22 AM, and 1/22 PM), the kitchen fan was turned on to depressurize the building and increase the air exchange rate. The HVAC system was also manipulated to change indoor/outdoor temperature difference, in addition to either no, continual, or intermittent HVAC fan operation. The best fit parameters obtained by the physical model and used in Figure 1 are shown in Table 1.

As Figure 1 and the R squared values in Table 1 show, the simple 2 parameter model is not adequate to describe all of the variability in the indoor concentration. Of the three chemical constituents studied, sulfate particles are arguably the most chemically stable.

Table 1: Model fits for data from January 16-23, 2001.

	PM 2.5 Sulfate	PM 2.5 Carbon	PM 2.5 Nitrate
Penetration Factor	0.95	1.03	0.67
Deposition Loss Rate (/h)	0.19	1.34	2.57
R squared	0.63	0.51	0.70

The stability of the PM2.5 sulfate is reflected by the fact that the penetration factor and deposition loss rate for sulfate are consistent with those expected for submicron particles. The carbon measured by the ICVC system includes both organic and inorganic carbon. For this total carbon fraction, there may be additional loss mechanisms, such as degradation or volatilization indoors. These additional loss terms are not distinguished in the model from the deposition loss rate term and thus will lead to the apparently high deposition losses. For nitrate, volatilization of ammonium nitrate into gaseous ammonia and nitric acid, causes a significant loss of particle nitrate in the indoor environment. Volatilization appears to account for a significant portion of the difference in penetration and deposition loss rates between nitrate and sulfate particles.

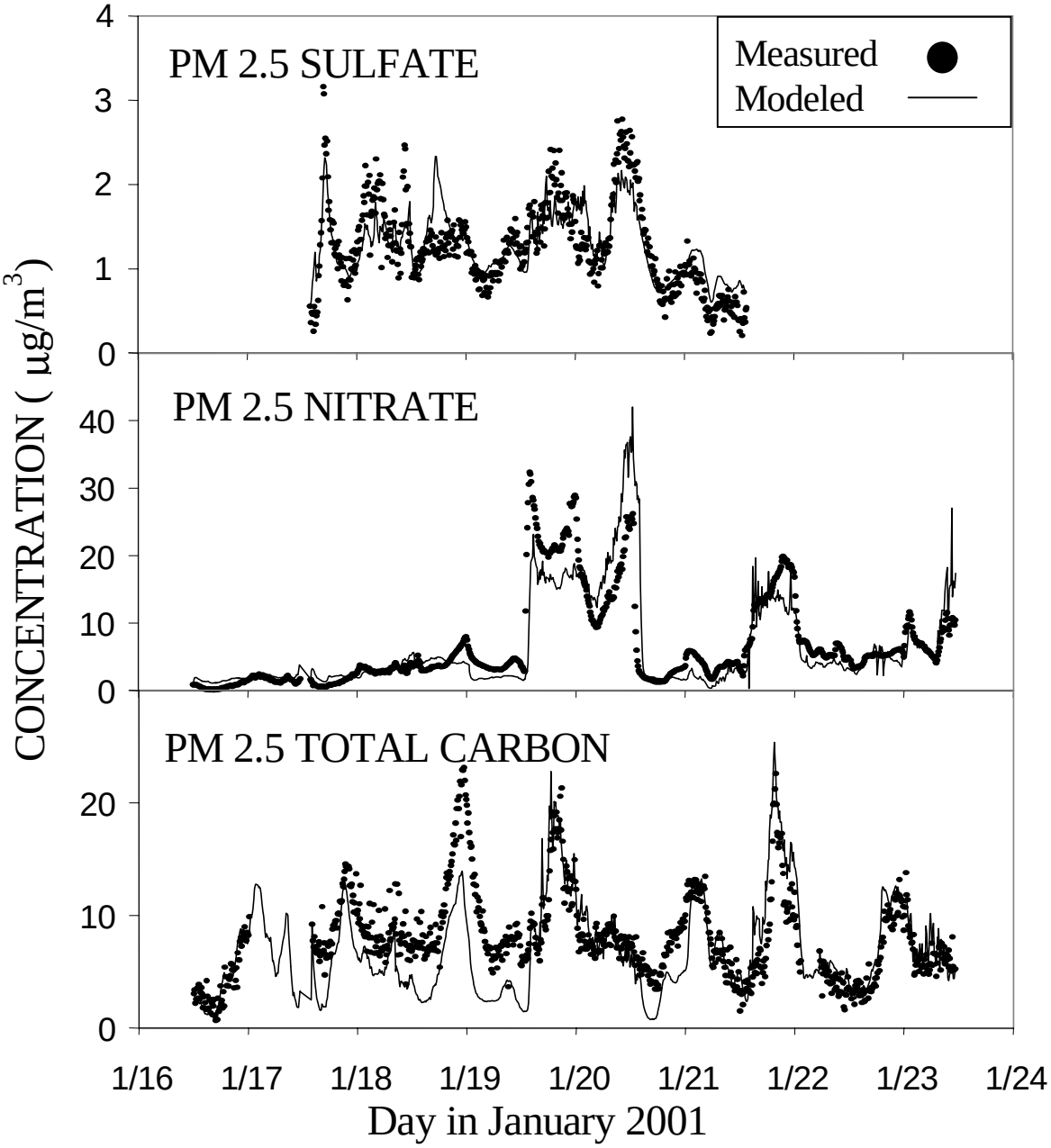

Figure 1: Comparison between the measured and modeled indoor concentrations using equation 2 and the parameters shown in Table 1. Tick marks on the x axis indicate midnight.

A multivariable linear regression was performed to investigate possible causes for the discrepancy between measured and modeled concentrations. The difference between the measured and modeled indoor concentration was compared to four variables:(1) outdoor minus indoor temperature, (2) outdoor minus indoor relative humidity, (3) infiltration rate, and (4) outdoor concentration of the compound. While we do not necessarily expect the form of the relationship between these variables and indoor concentration to be linear, this analysis can be used to explore whether a variable causes a significant effect and thus warrants further study. Table 2 shows the results of the regression. An example of the relationship is shown in Figure 2 where a definite trend is observed, despite considerable scatter in the data.

Table 2: Coefficients and standard errors from the four variable linear regression analysis for the three compounds measured, along with the R squared value for the regression. Coefficients with P [0.05 are shown in bold.

	PM 2.5 Sulfate	**PM 2.5 Carbon**	**PM 2.5 Nitrate**
T(out) – T (in)	**0.015 ± 0.005**	-0.026 ± 0.026	0.007 ± 0.033
%RH(out) – %RH(in)	**0.085 ± 0.008**	**0.474 ± 0.046**	**-0.661 ± 0.058**
Infiltration Rate (/h)	**0.073 ± 0.011**	**-1.009 ± 0.050**	**-0.444 ± 0.070**
C (out) (ug/m3)	**-0.110 ± 0.027**	-0.021 ± 0.011	**-0.059 ± 0.012**
R squared	0.24	0.44	0.28

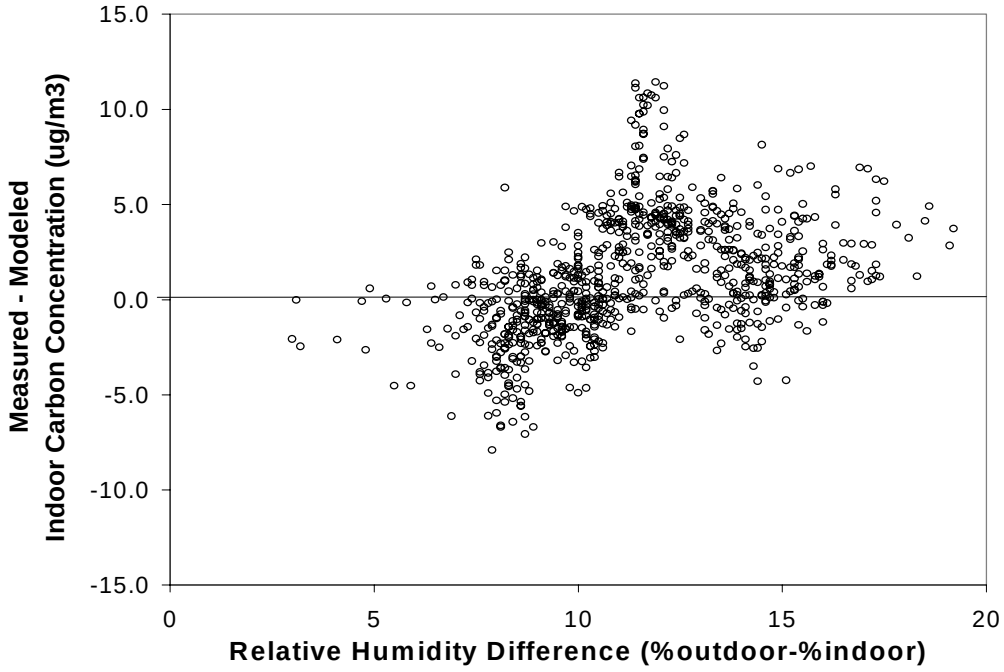

Figure 2: Indoor PM2.5 carbon model fit (measured - modeled concentration) versus relative humidity difference (%RH outdoor - % RH indoor).

The R squared values for the regressions suggest that a substantial fraction of the model/measured difference can be attributed to the factors tested, with the greatest influence found for total carbon. The indoor/outdoor temperature difference showed little or no correlation with the model fit for any of the chemical compounds studied. Both relative humidity difference and infiltration rate showed a significant correlation for all three particle

types, although the sign of the correlation varied between compounds. There was a significant correlation with outdoor concentration for sulfate particles, a small but significant correlation for nitrate particles, and a statistically insignificant correlation for carbon particles.

CONCLUSIONS

A time-dependent physical model was used to model indoor concentrations of three components of outdoor PM 2.5: carbon, nitrate, and sulfate. The model was shown to provide reasonable fits to the data over time periods of several days. However, for some time periods the measured indoor concentrations varied significantly from the model values. A multivariable linear regression was used to investigate possible causes for the model/measurement discrepancy. The four variables tested accounted for less than half of the model-measurement discrepancy observed. This indicates that there are likely to be other significant factors which have not been investigated in this study and/or that some effects are non-linear. Possible factors which may cause shifts in the deposition and penetration rates include changes in indoor flow conditions and shifts in the PM 2.5 size distribution for a chemical compound. Changes in wind speed and/or direction could also lead to shifts in the penetration rate if the crack size distribution was not uniformly distributed around the building shell. PM 2.5 carbon may undergo evaporation and/or chemical reaction and shifts in the composition of PM 2.5 carbon could change the rates of these processes. PM 2.5 nitrate, in the form of ammonium nitrate, is highly volatile in the indoor environment and the gaseous composition of the aerosol (ammonia and nitric acid) may affect the rate of volatilization. Considerable work will be required to determine which factors are most important in describing indoor concentrations of specific chemical compounds in outdoor PM 2.5.

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